QUANTUM-STATISTICAL THEORY OF RAMAN SCATTERING PROCESSES

A. MIRANOWICZ

and

S. KIELICH

Nonlinear Optics Division, Institute of Physics, Adam Mickiewicz University, Poznań, Poland

CONTENTS

I. INTRODUCTION: HISTORICAL DEVELOPMENTS

Almost simultaneously in 1928 Raman and Krishnan[1, 2] and Landsberg and Mandel'stamm[3] observed a new kind of scattering, now referred to as

Modern Nonlinear Optics, Part 3, Edited by Myron Evans and Stanisław Kielich. Advances in Chemical Physics Series, Vol. LXXXV.
ISBN 0-471-30499-9

(spontaneous) Raman scattering. For the last 65 years Raman scattering has unceasingly been in the forefront of both scientific and experimental investigations, particularly after the first observation of stimulated Raman scattering by Woodbury and Ng[4] (see also Ref. 5). Without exaggeration one can say that Raman scattering and spectroscopy constitute a completely autonomous discipline.

The literature on Raman scattering is quite prodigious. The theoretical principles and milestone experiments describing the Raman effect are summarized in a number of excellent monographs and reviews, for instance, by Bloembergen,[6] Kaiser and Maier,[7] Koningstein,[8] Grasyuk,[9, 10] Wang,[11, 12] Cardona,[13] Long,[14] Hayes and Loudon,[15] Penzkofer et al.,[16] Kielich,[17-19] Shen,[20] D'yakov and Nikitin,[21] and the most recent reviews by Raymer and Walmsley,[22] Peřina,[23] and Mostowski and Raymer.[24] We also refer the reader to the special issue of the *Journal of the Optical Society of America B*,[25] which is devoted entirely to Raman scattering. Although an extensive literature has accumulated dealing with Raman scattering, it should be emphasized that the understanding of the fundamental principles that govern the process is still incomplete.

There are several major groups of theories treating the Raman effect in the semiclassical and quantum approaches, and theories for standing waves and spatially propagating waves. Here, we discuss in detail the quantum theory of Raman scattering for several radiation modes only; this implies that the theory is the best suited for scattering in a tuned cavity. Nevertheless, some predictions from the standing wave model also can be applied for traveling wave models.[26-29]

Various methods have been applied to the Raman effect in each of the above theories. Taking into account the equation of motion as the basis for classification, we can distinguish the following approaches, based on the photon rate equation, the Schrödinger equation, the Heisenberg equation (Heisenberg-Langevin equation), the master equation (generalized Fokker-Planck equation), and the Maxwell-Heisenberg equation (Maxwell-Block equation); we refer to Refs. 22, 23, 30, 31. The above classification is obviously oversimplified. Firstly, there are many relations bridging these approaches. For instance, we shall apply the master equation approach from which we shall derive the Fokker-Planck equation and the photon rate equation. Secondly, there exist other alternative methods, which do not fit into our classification. Let us mention, for example, those developed by Mavroyannis[32-34] and Freedhoff.[35] Thirdly, one can classify the Raman effect theories in many other ways (see, e.g., Ref. 22).

We shall be considering the incident laser photons to be scattered by chaotic phonons or quantized chaotic vibrations in a crystal. The process leads to Stokes and anti-Stokes photons. To the description of Raman scattering, we use two trilinear Hamiltonians coupled via an infinite

number of phonon modes; one Hamiltonian describes Stokes radiation, and the other describes anti-Stokes radiation. The problem of coupled Stokes and anti-Stokes modes were studied previously by Bloembergen and Shen,[36-38] who applied the coupled wave theory of nonlinear optics formulated by Armstrong et al.[39] Later, Mishkin and Walls[40] quantized the Stokes and anti-Stokes modes, but dealt with the laser mode as a constant amplitude (so-called the parametric approximation). In fact, they considered two bilinear Hamiltonians, coupled by way of a phonon mode. Stokes scattering was treated as a parametric amplifier, whereas anti-Stokes scattering was treated as a parametric frequency converter. A detailed study of quantum statistics of the bilinear Hamiltonians, proposed by Louisell et al.,[41] has been extensively carried out (e.g., Refs. 42–50) and applied to Raman scattering, in particular by Mishkin and Walls,[40] Walls,[51, 52] Peřina,[53-55] Kárská and Peřina,[56] and others. Walls[57] (see also Ref. 44) has extended the bilinear Hamiltonian to a trilinear form to describe Raman scattering. The dynamics of Raman processes with trilinear Hamiltonian has been studied by Szlachetka et al.,[58-60] Szlachetka and Kielich,[61] Szlachetka,[27] Trung and Schütte,[62] Tänzler and Schütte,[63] Reis and Sharma,[64] Peřina et al.,[65, 66] Peřina and Křepelka,[67, 68] Levenson et al.,[69] and others (for general analyses see also Refs. 23, 70, and 71).

We shall describe Raman scattering from phonons as collective phenomena involving the interaction of many molecules. Much attention has also been drawn to a microscopic picture of the Raman effect by considering the interaction with individual molecules. Shen,[26, 72] in his quantum-statistical theory of nonlinear phenomena, proposed the general $m + n$ photon Hamiltonian, describing m emissions and n absorptions, and atomic transitions of an ensemble of N f-level atoms. This microscopically correct Hamiltonian contains Bose operators of a field and Fermi operators for optically active electrons and therefore describes a variety of nonlinear phenomena, in particular Raman scattering (for two- or three-level atoms). The same general Hamiltonian has been used by Walls[52] and McNeil and Walls.[73] Raman scattering from a two-level molecular (atomic) system[74-84] and in a three-level molecular system[74, 85-92] has been extensively studied by various authors. Walls[74] has shown that a description of Raman scattering from two-level molecules with a large cooperation number (coherent molecular coupling) is markedly similar to the results for Raman scattering from phonons. This is because for coherent molecular coupling, sums of the Fermi operators for the individual molecules can be replaced by the collective operators approximately satisfying boson commutation relations.

Nonclassical properties of radiation, such as squeezing, sub-Poissonian photon statistics, and photon antibunching, remain central topics in quantum optics. The literature in this area is truly prodigious. The reader is

referred to the articles published in Vol. 85 of this series and references therein, for instance, Refs. 93–95, as well as the reviews by Kielich et al.,[96] Leuchs,[97] Loudon and Knight,[98] Teich and Saleh,[99, 100] Zaheer and Zubairy,[101] and the topical issues of the *Journal of the Optical Society of America B*[102] and the *Journal of Modern Optics*.[103]

Squeezing properties of Raman scattering have been studied by Peřinová et al.,[65, 66] Peřina,[55] Kárská and Peřina,[56] Levenson et al.,[69] and Peřina and Křepelka.[67, 68] Sub-Poissonian photon-counting statistics and/or photon anticorrelations (in particular antibunching) have been investigated within various approaches to the standing-wave Raman effect by Loudon,[30] Simaan,[75] Agarwal and Jha,[87] Trung and Schütte,[62] Szlachetka and Kielich,[61] Szlachetka et al.,[58–60] Gupta and Mohanty,[78, 79] Peřina,[53, 54] Tänzler and Schütte,[63] Germey et al.,[104] Mohanty et al.,[80] Král,[105] Gupta and Dash,[90, 106] Ritsch et al.,[92] and in papers already mentioned[55, 56, 65–68] (see also Ref. 23). We note that photons scattered in the hyper-Raman effect can also exhibit nonclassical photon-counting correlations[27, 60, 65, 106–116] or squeezing.[65, 113, 116]

We shall analyze, in particular, cross-fluctuations (cross-correlations) in quadratures and in photocount statistics between different radiation modes. The theory of coherent light scattering within the consistent multipole tensor formalism, developed by Kielich[117] (see also Refs. 27, 58, 60) was successfully applied to disclose a novel cross-fluctuation mechanism. Here, an analysis of cross-correlations is presented along the lines of Szlachetka et al.[58, 59] (see also Ref. 23), as well as Loudon.[30]

We shall be studying sub- or super-Poissonian photon-counting statistics. We shall not analyze photon antibunching or bunching. The inclusion, in our Raman scattering model, of standard (i.e., temporal) photon antibunching would pose no problem. Let us mention the difference between sub-Poissonian statistics and anti-bunching pointed out in Refs. 118 and 119, which enables us to claim that these are distinct phenomena, and definitions should not be confused. The Raman scattering model is not suitable for investigations of spatial antibunching as defined by Le Berre-Rousseau et al.[120] and Białynicka-Birula et al.[121] in terms of negative angular correlations of photons.

As stated above, we shall be considering the quantum statistics of Raman scattering from phonons. We shall concentrate on a statistical analysis within the master equation approach to the Raman effect proposed by Shen,[26] Walls,[122] and McNeil and Walls.[73] This approach has been studied by various authors, e.g., Simaan,[75] Schenzle and Brand,[31] Peřina,[23, 53, 54] Germey et al.,[104] Gupta and Dash,[106] Bogolubov et al.,[88] Grygiel,[83] Miranowicz,[84] and Kárská and Peřina.[56] Usually a master equation is converted to a classical differential equation. Here, we shall apply a

transformation to a Fokker-Planck equation (FPE) for s-parametrized quasidistributions using the coherent-state technique and an alternative method of a master equation in terms of Fock states (or a rate equation for the conditional photon-number probabilities).

Walls[122] was the first to apply the FPE technique to Raman scattering. This approach was extensively developed by Peřina and coworkers (Ref. 23 and references therein). Unfortunately, a FPE for the Raman effect has been solved exactly under parametric approximation only; i.e., a pump depletion was not included. It means that Raman scattering is described as a competing process of parametric amplification (Stokes scattering) and parametric frequency conversion of light (anti-Stokes scattering) in a nonlinear crystal. This approximation seems to be a real shortcoming of the FPE approach. A problem of the existence of a solution of the FPE also arises. A diffusion matrix of the FPE for the s-parametrized quasidistributions (with $s \approx 1$) in many cases is not positive or positive semidefinite. Therefore, such a FPE cannot be interpreted as an equation of motion describing the Brownian motion under the influence of a suitable force.[123] For this reason the term pseudo- or generalized-FPE is used in the literature. It is sometimes argued that equations of this type are unphysical. However by doubling the phase space, it is possible to introduce a generalized P-representation (the positive P-representation).[124, 125] The equation of motion for this generalized P-representation is a FPE with a positive or positive semidefinite diffusion matrix. The nonpositive definite diffusion matrix plays an essential role in the production of nonclassical fields.[126]

The second method of an equation of motion in Fock representation has been applied to various multi-photon Raman processes.[30, 73, 75, 78–80, 90, 106, 107, 112, 114–116] The master equation (in terms of Fock states) for first-order Stokes scattering can be solved by applying the Laplace transform method. Solutions obtained by McNeil and Walls,[73] Simaan,[75] and others apply only to the diagonal elements of the density matrix $\hat{\rho}$, which is a serious drawback of these formulations.[23] The photocount statistics (sub-Poissonian photon statistics, antibunching, or anticorrelation) can be fully analyzed using the diagonal, in Fock representation, matrix elements of $\hat{\rho}$ only. However, the phase properties of the fields,[129–131] or squeezing properties[98, 132] (which are sensitive to the phase of the field) require the availability of the non-diagonal terms of the density matrix. We shall derive, for Raman scattering including depletion of the pump field, an exact solution of the master equation for the complete density matrix in Fock representation $\langle n, m|\rho|n', m'\rangle$ with arbitrary n, m, n', m'.

The classical description of Raman scattering into both the Stokes and anti-Stokes fields seems to be well understood,[6, 20] contrary to quan-

tum description, which is hampered by the complexity of the underlying Hamiltonians and hence the complex structure of the equations of motion. One of the simplest nontrivial models describing the coupling of the Stokes and anti-Stokes scattering was proposed by Knight.[133] This Raman-coupled model, in which a single atom is coupled to a single-cavity mode by Raman type transitions, has attracted some attention and has been generalized to much more realistic experimental conditions in the subsequent papers by Phoenix and Knight,[134] Schoendorff and Risken,[135] Agarwal and Puri,[136, 137] as well as Gerry and Eberly,[138] Gerry,[139] Gerry and Huang,[140] and Gangopadhyay and Ray.[141] It is quite remarkable that there exists a strict *operator* solution[136, 137] of the master equation describing the evolution of the generalized Knight model, which describes the system of an atom undergoing Raman transitions between two degenerate levels on interaction with a quantized field in a lossy cavity driven by an external field including the effects of atomic dephasing collisions. An extension of the propagation theory of Raman effect[142, 143] to include anti-Stokes scattering has been developed by Kilin[144] and independently by Li et al.[145] As mentioned, we shall analyze another model of the Stokes-anti-Stokes coupling within the framework of the temporal theory of Raman effect proposed by Walls[122] and extensively studied by Peřina and coworkers (see Refs. 23 and 68).

We shall discuss only temporal variations of fields instead of full temporal and spatial analysis. The assumption of monochromatic pump, Stokes, and anti-Stokes fields restricts the validity of our theory to a cavity problem. However, a temporal evolution in a cavity problem can usually be converted to a corresponding steady-state propagation in a dispersionless medium by simply replacing the time variable t by $-z/c$, a "normalized" space variable z. This procedure permits us to address nonlinear optical phenomena, in particular Raman scattering, in a manner analogous to their classical treatment.[20, 26, 28, 29] Formal space–time analogies have also been pointed out in the differential equations for the propagation of short light pulses.[146] Obviously, a full quantum space–time description is considerably more complex and resides in solving equations of motion for an infinite number of creation (annihilation) operators of the single-mode radiation fields. The total spatially dependent field is a sum of the single-mode solutions.

Here we mention only some spatial propagation theories of Raman scattering. For a detailed analysis we refer the reader to the review by Raymer and Walmsley[22] and references therein. The temporal and spatial evolution of the radiation fields (laser, Stokes, and anti-Stokes fields) in Raman scattering was successfully described within the framework of the classical coupled wave theory developed by Bloembergen and Shen[36–38]

(see also Ref. 20). The first quantum theories of Raman scattering including spatial propagation were proposed independently by von Foerster and Glauber[147] and Akhmanov et al.[148] using the analogy of Raman effect and optical parametric amplification processes. Another method was proposed by Emel'yanov and Seminogov[149] and Mostowski and Raymer[142, 143] using the analogy between Raman scattering and superfluorescent processes.[150] Spectacular predictions of the latter theory have been, in particular, macroscopic pulse-energy fluctuations of the emitted radiation in a manner reflecting the underlying spontaneous initiation.[151-153] The negative-exponential probability distribution (NEPD), derived by Raymer et al.,[151] describes the macroscopic fluctuations of the scattered radiation. Kárská and Peřina[56] pointed out that the NEPD corresponds to the generating function of the integrated intensity extensively used in this paper (see Section V.B). The standing-wave theory of Raman scattering properly describes the macroscopic fluctuations in the low-gain and high-gain regime (see Refs. 22, 23, and 145 and references therein). In the latter limit the quantum fluctuations of the generated fields can be thought of as arising from a classical noise process, contrary to the low-gain limit, where certain nonclassical effects occur.

Finally, we should mention certain crucial experiments revealing some manifestations of Raman scattering. For more details, see the review article by Raymer and Walmsley.[22] Experiments on the detection of fluctuations of Stokes pulse energies were carried out by Walmsley and Raymer[154, 155] and Fabricius et al.[156] The temporal and spatial fluctuations of the Stokes beam profile, the spectrum, and delay have been investigated in a number of experiments both for depleted and undepleted pump pulse (for references see Ref. 22). We mention these experiments because the theory of Raman scattering for cavity modes, to be presented here, correctly predicts the existence of macroscopic quantum fluctuations of the Stokes pulses.

Generation of Raman solitons in the heavily depleted pump pulse has recently been observed by MacPherson et al.[157] and Swanson et al.,[158] as predicted by Englund and Bowden.[159, 160] Cooperative effects in Raman scattering, referred to as cooperative Raman scattering, which is analogous to two-level superfluorescence,[161] occurs for a laser pump not significantly depleted. The effect was first observed by Kirin et al.[162] and then re-examined under fully convincing experimental conditions by Pivtsov et al.[163] In our analysis we clearly distinguish the two cases of the depleted and undepleted pump field, and therefore have listed some effects and experiments in which this condition for the intensity pump is crucial.

Hyper-Raman scattering, i.e., the three-photon analog of Raman scattering, was discovered in 1965 by Terhune et al.[164] (see also Réf. 165). This

effect was predicted theoretically by Neugebauer,[166] Kielich,[167, 168] and Li[169] prior to its experimental detection. Since the discovery of hyper-Raman scattering, numerous papers have appeared reporting theoretical investigations and observations of the process in a variety of solids, liquids and vapours. Here, we shall not discuss higher-order Raman scattering processes. We refer to the reviews of Refs. 17, 23, 27, and 172 for details and literature.

This paper is organized as follows. In Section II, the standing-wave model of Raman scattering is constructed and the basic equation of motion (master equation) is derived. In Section III, we give a short account of multimode s-parametrized quasidistributions and s-parametrized characteristic functions. In Section IV, we introduce definitions of nonclassical properties of radiation such as quadrature ("usual" and principal) squeezing and sub-Poissonian photon statistics. In Section V, we present the s-parametrized quasidistribution formalism of Raman scattering either including (in Section V.A) or neglecting (in Section V.B) depletion of the pump laser beam in the process of scattering. In Section VI we develop the density matrix formalism of Raman scattering. We derive exact solutions of the master equation in Fock representation in Section VI.A.2. We also give short-time (in Section VI.A.1) and long-time (in Section VI.A.3) solutions of the master equation. In Section VI.B we present approximate solutions valid under parametric approximation, i.e., when pump depletion is neglected.

II. MODEL AND MASTER EQUATION

Let us analyze Raman scattering starting from a completely quantum Hamiltonian but describing phenomenologically only the net effect, i.e., ignoring the details of the scattering mechanism. We describe the interaction of three single-mode radiation fields: an incident laser beam at the frequency ω_L, a Stokes field at the frequency ω_S, and an anti-Stokes field at the frequency ω_A through an infinite phonon system at frequencies ω_{V_j}, after Walls[122] (see also Refs. 53–55), by the effective Hamiltonians:

$$\hat{H}_0 = \hbar\omega_L \hat{a}_L^+ \hat{a}_L + \hbar\omega_S \hat{a}_S^+ \hat{a}_S + \hbar\omega_A \hat{a}_A^+ \hat{a}_A + \hbar \sum_j \omega_{V_j} \hat{a}_{V_j}^+ \hat{a}_{V_j} \tag{1}$$

$$\hat{H}_S = \hbar \sum_j \lambda_{S_j} \hat{a}_L \hat{a}_S^+ \hat{a}_{V_j}^+ + \text{h.c.}$$

$$\hat{H}_A = \hbar \sum_j \lambda_{A_j}^* \hat{a}_L \hat{a}_A^+ \hat{a}_{V_j} + \text{h.c.} \tag{2}$$

$$\hat{H}_T = \hat{H}_0 - \hat{H}_S - \hat{H}_A \tag{3}$$

where $\hat{H}_S$ ($\hat{H}_A$) is the trilinear interaction Hamiltonian for Stokes (anti-Stokes) scattering and H_0 is the unperturbed Hamiltonian. For simplicity, we have dropped the zero-point contributions. The annihilation operators for the laser, Stokes, anti-Stokes, and phonon fields are denoted by $\hat{a}_L$, $\hat{a}_S$, $\hat{a}_A$, and $\hat{a}_{Vi}$, respectively (we label all Hilbert space operators with caret). The coupling coefficient $\lambda_{Sj}(\lambda_{Aj})$ denotes the strength of the coupling between the Stokes (anti-Stokes) mode and the optical phonon at the frequency ω_{Vj}. These coefficients depend on the actual interaction mechanism. In the Hamiltonians (2) we neglect terms describing higher-order Stokes scattering,[57, 64, 74] as well as terms describing hyper-Raman scattering.[27, 65, 106] In Section VI.A in the analysis of the Raman effect without parametric approximation we also neglect anti-Stokes production.

In our model we take into account only the radiation modes appropriate for a cavity. It should be kept in mind that the several radiation mode description is applied to the waves involved in the whole course of the interaction, not only at the beginning of the interaction process. This approximation is a shortcoming from the experimental point of view, since it is not very suitable for describing the most common experimental arrangements used when measuring stimulated Raman scattering.[20-22]

We apply the rotating wave approximation since in the interaction Hamiltonians (2) we have omitted terms of the form $\hat{a}_{V_j}\hat{a}_S^+\hat{a}_L$ + h.c. and $\hat{a}_{V_j}^+\hat{a}_A^+\hat{a}_L$ + h.c.. For weak coupling these terms are negligible because they vary rapidly as $\exp[\pm i(\omega_{Vj} + |\omega_L - \omega_{S,A}|)t]$, which implies that their average is approximately zero for times of evolution much greater than $|\omega_L - \omega_{S,A}|^{-1}$, contrary to the interaction Hamiltonians H_S and H_A (2), which vary as $\exp[\pm i(\omega_{Vj} - |\omega_L - \omega_{S,A}|)t]$ giving unity for $\omega_{Vj} \approx |\omega_L - \omega_{S,A}|$. We have also neglected terms of the form $\hat{a}_{Vj}^{\pm}\hat{a}_S\hat{a}_L$ + h.c. and $\hat{a}_{Vj}^{\pm}\hat{a}_A\hat{a}_L$ + h.c.. These terms, if included, would describe a process in which both the Stokes (anti-Stokes) and laser photons are annihilated and created in the scattering act.

The Hamiltonians (2) describe Raman scattering under the long wavelength approximation, which has several important implications.[147, 173, 174] Firstly, we can neglect the intermolecular interactions. Each optical vibrational mode of the medium is equivalent to a simple harmonic oscillator. Secondly, the optical phonon dispersion is negligible. A typical dispersion curve for optical phonons, $\omega_V(k_V)$, is almost flat for wave vectors k_V from the interval $(-1/\lambda, 1/\lambda)$, where λ is an optical wavelength. In other words, optical wave vectors occupy only a very small volume about the origin of the reciprocal lattice. Thirdly, a crystal can be treated as a continuum; thus, from the mathematical point of view, sums over lattice sites can be replaced by integrals over a volume of the crystal. This long wave approximation is quite realistic for optical processes, in particular Raman scattering.

A detailed derivation of the Hamiltonian from first principles has been given by von Foerster and Glauber[147] in their quantum propagation theory of Raman scattering from phonons. Although we deal with modes in a cavity, many aspects of their theory recur in our approach.

In the case of an unbounded medium the momentum is conserved in the interaction, i.e., the sum (difference) of the wave vectors $\mathbf{k}_S$ ($\mathbf{k}_A$) of the Stokes (anti-Stokes) photon and $\mathbf{k}_V$ of the photon involved in the scattering act is exactly equal to the laser light wave vector $\mathbf{k}_L$,

$$\mathbf{k}_L = \mathbf{k}_S + \mathbf{k}_V \qquad \mathbf{k}_L = \mathbf{k}_A - \mathbf{k}_V \tag{4}$$

This means that each laser mode interacts strongly only with phonons having a single wave vector (one and only one vibrational mode). This is the requirement of translational invariance. Momentum is no longer strictly conserved for interactions in a finite medium, since the introduction of boundaries destroys the translational invariance of the medium. The strongest interaction is still for those modes which conserve momentum (4) and energy ($\omega_{Vj} \approx |\omega_L - \omega_{S,A}|$); nevertheless, in this case the radiation modes are coupled to a certain range of optical phonons whose wave vectors may not satisfy the adequate conditions (4) by amounts of the order of the reciprocal of the dimensions of the medium.[147] The coupling constants λ_{Sj}, and λ_{Aj} contain these momentum mismatches via phase integrals[27, 122]:

$$\begin{aligned} \lambda_{Sj} &\sim \int_V \exp\left[-\mathrm{i}(\mathbf{k}_L - \mathbf{k}_S - \mathbf{k}_{Vj}) \cdot \mathbf{r}\right] \mathrm{d}^3 r \\ \lambda_{Aj} &\sim \int_V \exp\left[-\mathrm{i}(\mathbf{k}_L - \mathbf{k}_A + \mathbf{k}_{Vj}) \cdot \mathbf{r}\right] \mathrm{d}^3 r \end{aligned} \tag{5}$$

Hence, the interaction Hamiltonians (2) are represented by sums over all optical vibrational modes that may scatter into or out of the desired mode. This means that the coupling of the radiation fields (in particular Stokes and anti-Stokes) through a large number of optical phonons is treated stochastically.

In the Hamiltonians (1)–(3) we have assumed all the radiation fields and phonons to be polarized linearly in the same direction. We have not included explicitly the polarization states of those photons, which might

affect the photon-counting statistics,[175, 176] squeezing,[177, 178] and other properties (Ref. 179 and references therein). Obviously, this would require the discussion of correlation tensors, in place of correlation functions, involving the photon polarization states.[19, 180, 181]

The model under discussion is restricted to the approximation of electric-dipole transitions. In previous papers,[170, 171] Kielich has proposed and extensively developed the formal quantum theory of first-, second-, and higher-order processes (in particular Raman scattering) taking into account multipolar electric and magnetic quantum transitions.

A lot of attention has been devoted to a simpler completely boson Hamiltonian applied to the description of the statistical properties of Raman scattering by phonons treated as a single monochromatic mode (Refs. 23, 44, 57, 58, and 69 and references therein). It is clear that the use of a large number of phonon modes (a phonon bath) in the model Hamiltonians (2) provides a fuller picture of the scattering processes. In particular, the model describes the stochastic coupling of the Stokes and anti-Stokes modes through a phonon bath. The assumption of a single phonon mode implies that the Stokes and anti-Stokes fields are coupled in a deterministic manner, which seems to be a rather serious drawback.[122]

As a digression, let us mention that the same phenomenological Hamiltonians (2) have been used in the description of Brillouin scattering (see, for example, Refs. 23, 104, and 182 and references therein). The main difference between Brillouin and Raman scattering lies in different kinds of the scatterers responsible for these effects: acoustic phonons in the Brillouin effect, and optical phonons in the Raman effect. This difference is included in the frequencies, the coupling constants $\lambda_{Sj}, \lambda_{Aj}$, and the reservoir spectrum. More important, acoustic phonons exhibit much greater dispersion than optical phonons. In our approach to Raman scattering we neglect dispersion. This assumption applied to Brillouin scattering has considerably less validity.

We are interested only in the statistical properties of the radiation fields (the pump and scattered beams) considered as a system. We therefore remove the unnecessary information about the infinite system of optical phonons, treated as a reservoir (heat bath). The procedure leading to the master equation is widely used in quantum optics. For a general review of the master equation methods and the extensive bibliography see Refs. 23, 51, 183, 184. We rewrite the interaction Hamiltonians H_S and H_A in the interaction picture as

$$\hat{H}_S + \hat{H}_A = \hbar \sum_{k=1}^{4} \hat{F}_k \hat{Q}_k \tag{6}$$

where

$$\begin{aligned}
\hat{F}_1 = \hat{F}_2^+ &= \sum_j \lambda_{Sj} \hat{a}_{Vj}^+ \exp\left[\mathrm{i}\omega_{Vj}(t - t_0)\right] \\
\hat{F}_3 = \hat{F}_4^+ &= \sum_j \lambda_{Aj}^* \hat{a}_{Vj} \exp\left[-\mathrm{i}\omega_{Vj}(t - t_0)\right] \\
\hat{Q}_1 = \hat{Q}_2^+ &= \hat{a}_L \hat{a}_S^+ \exp\left[-\mathrm{i}\Omega_S(t - t_0)\right] \\
\hat{Q}_3 = \hat{Q}_4^+ &= \hat{a}_L \hat{a}_A^+ \exp\left[\mathrm{i}\Omega_A(t - t_0)\right]
\end{aligned} \tag{7}$$

The $\hat{Q}_i$ ($\hat{F}_i$) are respectively functions of the system (reservoir) operators only. The "cavity" frequencies Ω_S, Ω_A are equal to

$$\Omega_{S,A} = |\omega_L - \omega_{S,A}| \tag{8}$$

Since the system and the reservoir variables are mutually independent, as it follows from

$$\left[\hat{a}_i, \hat{a}_j^+\right] = \delta_{ij} \qquad \text{for } i, j = L, S, A, V_1, V_2, \ldots \tag{9}$$

we may trace, in standard manner, the complete density matrix over the reservoir leading to the reduced density matrix $\hat{\rho}(t)$. Obviously, we cannot obtain any reservoir averages from $\hat{\rho}(t)$. There are some Raman scattering models (e.g., Refs. 23, 104, and 147), where optical phonons are included in the system, whereas other crystal excitation modes, such as acoustical phonons, electric excitations, and other species of molecular vibrations, serve as a thermal reservoir.

The radiation fields are weakly coupled to the thermal reservoir. The anti-Stokes mode loses energy to the reservoir. The fluctuations in the reservoir also couple back into the system introducing noise from the reservoir. However, we apply the Markov approximation, a condition sufficient to ensure that energy that goes into the reservoir will not return to the radiation fields. This conclusion follows from the definition of the Markovian system as one that cannot develop memory—the future of the system is determined by the present and not its past.[183, 184] The importance of this assumption is sometimes stressed in the concept of a Schrödinger-Markov (or Heisenberg-Markov) picture, meaning the standard pictures under Markov approximation.[183] The importance of non-Markovian effects in Raman scattering has been recently studied by, e.g., Sugawara et al.[190] and Villaeys et al.[91] Obviously, the system operators $\hat{Q}_i$

obey the same commutation relations under this approximation as they did originally.

To obtain the equation of motion for the reduced density matrix $\hat{\rho}(t)$, one has to compute the reservoir spectral densities

$$\begin{aligned} \omega_{ij}^{+} &= \int_0^\infty \dot{e}^{i\omega_i\tau}\langle \hat{F}_i(\tau)\hat{F}_j\rangle_R \, d\tau \\ \omega_{ji}^{-} &= \int_0^\infty e^{i\omega_i\tau}\langle \hat{F}_j\hat{F}_i(\tau)\rangle_R \, d\tau \end{aligned} \tag{10}$$

where $\langle \ldots \rangle_R$ is the average over all reservoir operators; ω_i takes the values $\pm\Omega_{S,A}$. The infinite system of optical phonons is assumed to be densely spaced with the number of modes between ω_i and $\omega_i + d\omega_i$ equal to $g(\omega_i)d\omega_i$, so we may replace the sums over the optical vibrational modes by integrals

$$\sum_j (\ldots) \approx \int_0^\infty d\omega_j g(\omega_j)(\ldots) \tag{11}$$

Let us introduce two quantities: $\Delta\Omega$–the frequency mismatch and Ω–the medium "cavity" frequency, defined by

$$\begin{aligned} \Delta\Omega &= \frac{\Omega_S - \Omega_A}{2} \\ \Omega &= \frac{\Omega_S + \Omega_A}{2} \end{aligned} \tag{12}$$

The frequency mismatch $\Delta\Omega$, in general, is not equal to zero. It is quite realistic for optical phonons to assume that the coupling constants $\lambda_{S,A}(\omega_j)$ and the phonon density of $g(\omega_j)$ are flat in the vicinity of Ω, so that we can write

$$\begin{aligned} g(\Omega \pm \Delta\Omega) &\approx g(\Omega) \\ \lambda_k(\Omega \pm \Delta\Omega) &\approx \lambda_k(\Omega) \qquad k = S, A \end{aligned} \tag{13}$$

The reservoir is supposed to be at thermal equilibrium. The phonons are unaffected by interaction with the radiation fields. In the classical sense this means that the phonons are so quickly damped that they remain in their steady state.[20, 37, 38] The mean number of phonons in the reservoir

mode at thermal equilibrium is defined by the Bose-Einstein distribution

$$\langle \hat{n}(\omega_{V_j}) \rangle = \left[\exp\left(\frac{\hbar \omega_{V_j}}{k_B T} \right) - 1 \right]^{-1} \tag{14}$$

where k_B is the Boltzmann constant and T is the temperature of the reservoir. Obviously, as the reservoir temperature approaches absolute zero, the mean number of phonons $\langle \hat{n}(\omega_{V_j}) \rangle$ tends to zero as well. In Section V.B, we analyze Raman scattering in a parametric approximation for a "noisy" reservoir ($\langle \hat{n}(\omega_{V_j}) \rangle \neq 0$), whereas we study Raman scattering including the pump depletion for "quiet" reservoir ($\langle \hat{n}(\omega_{V_j}) \rangle \approx 0$). After some algebra one obtains from (10),

$$\begin{aligned}
\omega_{21}^{+} &= \left(\frac{\gamma_S}{2} + i\Delta\omega_S \right) \left(\langle \hat{n}_V \rangle + 1 \right) \\
\omega_{12}^{+} &= \left(\frac{\gamma_S}{2} - i\Delta\omega_S \right) \langle \hat{n}_V \rangle \\
\omega_{43}^{+} &= \left(\frac{\gamma_A}{2} - i\Delta\omega_A \right) \langle \hat{n}_V \rangle \\
\omega_{34}^{+} &= \left(\frac{\gamma_A}{2} + i\Delta\omega_A \right) \left(\langle \hat{n}_V \rangle + 1 \right) \\
\omega_{31}^{+} &= \left(\frac{\gamma_{AS}}{2} + i\Delta\omega_{AS} \right) \left(\langle \hat{n}_V \rangle + 1 \right) \\
\omega_{13}^{+} &= \left(\frac{\gamma_{SA}}{2} - i\Delta\omega_{SA} \right) \langle \hat{n}_V \rangle \\
\omega_{42}^{+} &= \left(\frac{\gamma_{AS}}{2} - i\Delta\omega_{AS} \right) \langle \hat{n}_V \rangle \\
\omega_{24}^{+} &= \left(\frac{\gamma_{SA}}{2} + i\Delta\omega_{SA} \right) \left(\langle \hat{n}_V \rangle + 1 \right) \\
\omega_{ij}^{-} &= \left(\omega_{ij}^{+} \right)^{*}
\end{aligned} \tag{15}$$

All other reservoir spectral densities, in particular the diagonal densities $\omega_{ii}^{\pm}$ (for $i = 1, \ldots, 4$), vanish. For simplicity we have denoted the mean number of phonons at frequency Ω by $\langle \hat{n}_V \rangle = \langle \hat{n}_V(\Omega) \rangle$. The gain constant for the Stokes mode γ_S, the damping constant for the anti-Stokes model γ_A, and the mutual damping constants for both scattered fields

γ_{SA}, γ_{AS} are

$$\begin{aligned} \gamma_k &= 2\pi g(\Omega)|\lambda_k(\Omega)|^2 \qquad (k = S, A) \\ \gamma_{SA} &= \gamma_{AS}^* = 2\pi g(\Omega)\lambda_S(\Omega)\lambda_A^*(\Omega) \end{aligned} \tag{16}$$

where $g(\Omega)$, as earlier, denotes the density of the optical phonon modes (the reservoir spectrum) at frequency Ω. It is seen that the following simple relation between the single and mutual damping constants holds: $|\gamma_{SA}|^2 = |\gamma_{AS}|^2 = \gamma_A\gamma_S$. The frequency shifts, representing the Lamb shift in the frequency $\Omega \approx \Omega_j$, are expressed by the Cauchy principle value, $\mathscr{P}$ of the integrals:

$$\begin{aligned} \Delta\omega_k &= -\mathscr{P}\int_0^\infty \frac{g(\omega_j)|\lambda_k(\omega_j)|^2}{\omega_j - \Omega_k}\,\mathrm{d}\omega_j \qquad (k = S, A) \\ \Delta\omega_{SA} &= (\Delta\omega_{AS})^* = -\mathscr{P}\int_0^\infty \frac{g(\omega_j)\lambda_S(\omega_j)\lambda_A^*(\omega_j)}{\omega_j - \Omega}\,\mathrm{d}\omega_j \end{aligned} \tag{17}$$

The only effect of the $\Delta\omega_i$ is to change slightly the frequency Ω, so we neglect them. Having calculated the reservoir spectral densities we can write the master equation for the reduced density matrix $\hat{\rho} = \hat{\rho}(\hat{a}_L, \hat{a}_S, \hat{a}_A, t)$ as

$$\begin{aligned} \frac{\partial}{\partial t}\hat{\rho} &= \tfrac{1}{2}\gamma_S\left(\left[\hat{a}_L\hat{a}_S^+, \hat{\rho}\hat{a}_L^+\hat{a}_S\right] + \text{h.c.}\right) \\ &\quad + \tfrac{1}{2}\gamma_A\left(\left[\hat{a}_L^+\hat{a}_A, \hat{\rho}\hat{a}_L\hat{a}_A^+\right] + \text{h.c.}\right) \\ &\quad + \tfrac{1}{2}\gamma_{SA}\,\mathrm{e}^{-2\mathrm{i}\Delta\Omega\,\Delta t}\left(\left[\hat{a}_L\hat{a}_S^+, \hat{\rho}\hat{a}_L\hat{a}_A^+\right] + \left[\hat{a}_L\hat{a}_S^+\hat{\rho}, \hat{a}_L\hat{a}_A^+\right]\right) \\ &\quad + \tfrac{1}{2}\gamma_{AS}\,\mathrm{e}^{2\mathrm{i}\Delta\Omega\,\Delta t}\left(\left[\hat{a}_L^+\hat{a}_A, \hat{\rho}\hat{a}_L^+\hat{a}_S\right] + \left[\hat{a}_L^+\hat{a}_A\hat{\rho}, \hat{a}_L^+\hat{a}_S\right]\right) \\ &\quad - \langle\hat{n}_V\rangle\Big\{\tfrac{1}{2}\gamma_S\left(\left[\hat{a}_L^+\hat{a}_S, \left[\hat{a}_L\hat{a}_S^+, \hat{\rho}\right]\right] + \text{h.c.}\right) \\ &\qquad + \tfrac{1}{2}\gamma_A\left(\left[\hat{a}_L\hat{a}_A^+, \left[\hat{a}_L^+\hat{a}_A, \hat{\rho}\right]\right] + \text{h.c.}\right) \\ &\qquad + \gamma_{SA}\,\mathrm{e}^{-2\mathrm{i}\Delta\Omega\,\Delta t}\left[\hat{a}_L^+\hat{a}_S, \left[\hat{a}_L^+\hat{a}_A, \hat{\rho}\right]\right] \\ &\qquad + \gamma_{AS}\,\mathrm{e}^{2\mathrm{i}\Delta\Omega\,\Delta t}\left[\hat{a}_L\hat{a}_A^+, \left[\hat{a}_L\hat{a}_S^+, \hat{\rho}\right]\right]\Big\} \end{aligned} \tag{18}$$

The term in γ_S represents the amplification of the Stokes mode; the term in γ_A describes the loss of energy from the anti-Stokes mode into the

reservoir; the γ_{AS} and γ_{SA} terms represent the stochastic coupling between the Stokes and anti-Stokes modes through the reservoir; the remaining terms in $\langle \hat{n}_V \rangle \gamma_i$ represent the diffusion of fluctuations of the reservoir into the system modes. Eq. (18) describes, moreover, the evolution of the laser beam, i.e., the depletion of the laser field, the coupling of the field with the Stokes and anti-Stokes fields, as well as the diffusion of the reservoir fluctuations into the laser field. The interpretation of the γ_S (γ_A) terms as the amplification (attenuation) of the radiation fields is as yet intuitive, but will gain in precision on solution of the generalized Fokker-Planck equation. The master equation (18) could have been written in more compact form; albeit for purposes of interpretation the above form is more convenient.

The master equation (18), in the particular case of parametric approximation, reduces to the equation obtained by Walls[122] and Peřina,[53] and reduces to that of McNeil and Walls for Stokes scattering alone but with no need for the parametric approximation.[73] Our master equation (18) differs but slightly in the diffusion terms $\langle \hat{n}_V \rangle \gamma_i$ only from the special case of the master equation given by Agarwal[185] (see also Ref. 73).

The master equation may be solved by various techniques presented in standard textbooks.[23, 183, 184, 186–188] Here, we apply two methods. We convert the master equation to an associated classical equation. On the one hand, expressing the quantum equation in s-ordered form one obtains the generalized Fokker-Planck equation for the s-parametrized quasi-probability distribution, which can be exactly solved for a class of Ornstein-Uhlenbeck processes.[188, 191] On the other hand, one can express the master equation in Fock representation, which can be solved, for instance, by the Laplace transform method.[30, 73, 75]

In the following sections we analyze three cases. Firstly, we briefly describe coupling of the three quantum radiation fields: the laser, Stokes, and anti-Stokes beams. The problem simplifies considerably if one assumes narrow quasi-probability distributions. Secondly, we apply the parametric approximation, which means that the pump field is treated classically. We include the coupling of the Stokes and anti-Stokes field through the phonon bath. Thirdly, we separately describe either the laser and Stokes mode or the laser and anti-Stokes mode, but include the depletion of the pump laser light. In this case we assume the heat bath to be "quiet."

III. MULTIMODE s-PARAMETRIZED QUASIDISTRIBUTIONS

A description of the multimode fields via quasiprobability distributions (quasidistributions, QPDs) or equivalently via characteristic functions was

first proposed by Glauber,[180, 192, 193] Cahill,[194] and Klauder et al.[195] General ordering theorems have been given by Agarwal and Wolf.[196] The s-parametrized single-mode quasidistributions and characteristic functions were introduced by Cahill and Glauber,[197] who extensively studied various ways of defining correspondences between the operators and functions. For a recent review of the multimode s-parametrized functional formalism we refer the reader to Ref. 23. Here, we list the basic definitions and properties of the s-parametrized multimode quasidistributions and characteristic functions useful for our further investigations.

To solve the master equation (18), i.e., the operator equation, we use the c-number representations $\mathscr{W}^{(s)}(\{\alpha_k\})$ and $\mathscr{C}^{(s)}(\{\beta_k\})$ of the density operator introduced by Cahill and Glauber.[197] These representations not only are useful as a calculation tool, but also provide insight into the interrelations between classical and quantum mechanics. By virtue of the multimode s-parametrized displacement operator

$$\hat{D}^{(s)}(\{\beta_k\}) = \Pi_k \hat{D}^{(s)}(\beta_k) = \prod_k \exp\left(\beta_k \hat{a}_k^+ - \beta_k^* \hat{a}_k + \frac{s}{2}|\beta_k|^2\right) \quad (19)$$

where the continuous parameter s belongs to the interval $\langle -1, 1\rangle$, one can define the s-parametrized multimode characteristic function as the mean value of $\hat{D}^{(s)}(\{\beta_k\})$,

$$\mathscr{C}^{(s)}(\{\beta_k\}) = \mathrm{Tr}\left[\hat{\rho}\hat{D}^{(s)}(\{\beta_k\})\right] \quad (20)$$

In our situation involving the three radiation modes laser ($k = L$), Stokes (S) and anti-Stokes (A), the simplified notation in Eqs. (19) and (20) stands for $(\{\beta_k\}) = (\beta_L, \beta_S, \beta_A)$. The Fourier transform of the characteristic function $\mathscr{C}^{(s)}(\{\beta_k\})$ (20) readily gives the s-parametrized multimode quasidistribution $\mathscr{W}^{(s)}(\{\alpha_k\})$,

$$\mathscr{W}^{(s)}(\{\alpha_k\}) = \int \mathscr{C}^{(s)}(\{\beta_k\}) \exp\left[\sum_k (\alpha_k \beta_k^* - \alpha_k^* \beta_k)\right] \mathrm{d}^2\left\{\frac{\beta_k}{\pi}\right\} \quad (21)$$

For completeness we write the inverse Fourier transform, which enables us to determine $\mathscr{C}^{(s)}$ from $\mathscr{W}^{(s)}$, namely,

$$\mathscr{C}^{(s)}(\{\beta_k\}) = \int \mathscr{W}^{(s)}(\{\alpha_k\}) \exp\left[\sum_k (\alpha_k^* \beta_k - \alpha_k \beta_k^*)\right] \mathrm{d}^2\left\{\frac{\alpha_k}{\pi}\right\} \quad (22)$$

where integration extends over α_k in the following sense:

$$d^2\left\{\frac{\alpha_k}{\pi}\right\} = \prod_{k=L,S,A} \pi^{-1}\, d^2\alpha_k = \pi^{-3} \prod_{k=L,S,A} d(\mathrm{Re}\,\alpha_k)\, d(\mathrm{Im}\,\alpha_k)$$

or over β_k similarly. The normalization is chosen to satisfy

$$\int \mathscr{W}^{(s)}(\{\alpha_k\})\, d^2\left\{\frac{\alpha_k}{\pi}\right\} = \mathscr{C}^{(s)}(0) = 1 \tag{23}$$

In the three special cases of $s = -1, 0, 1$ one recognizes the well-known QPDs,[23, 197, 201] namely the Q function, the Wigner function, and the Glauber-Sudarshan P-function, respectively:

$$\begin{aligned} Q(\{\alpha_k\}) &= \langle\{\alpha_k\}|\hat{\rho}|\{\alpha_k\}\rangle = \mathscr{W}^{(-1)}(\{\alpha_k\}) \\ W(\{\alpha_k\}) &= \mathscr{W}^{(0)}(\{\alpha_k\}) \\ P(\{\alpha_k\}) &= \pi^{-M}\mathscr{W}^{(1)}(\{\alpha_k\}) \end{aligned} \tag{24}$$

with M denoting the number of modes (in our analysis M will be equal to 3, 2, or 1). One can say that the s-parametrized quasidistribution $\mathscr{W}^{(s)}$ (with s from the interval $\langle -1, 1\rangle$) is a continuous interpolation between the P- and Q-functions. The Q-function directly determines antinormally ordered expectation values, the P-function determines normally ordered averages, and the Wigner function can be used directly to calculate the averages of symmetrically ordered operators. The following relations hold for any parameter s:

$$\begin{aligned} \Big\langle \prod_k (\hat{a}_k^+)^{m_k} (\hat{a}_k)^{n_k} \Big\rangle_{(s)} &= \mathrm{Tr}\left[\hat{\rho}\left\{\prod_k (\hat{a}_k^+)^{m_k} (\hat{a}_k)^{n_k}\right\}_{(s)}\right] \\ &= \int \mathscr{W}^{(s)}(\{\alpha_k\}) \prod_k (\alpha_k^*)^{m_k} (\alpha_k)^{n_k}\, d^2\{\alpha_k/\pi\} \\ &= \prod_k \frac{\partial^{m_k}}{\partial \beta_k^{m_k}} \frac{\partial^{n_k}}{\partial(-\beta_k^*)^{n_k}} \mathscr{C}^{(s)}(\{\beta_k\})\Big|_{\{\beta_k\}=0} \end{aligned} \tag{25}$$

where $\{\beta_k\} = 0$, in the three-mode case, means that $\beta_L = \beta_S = \beta_A = 0$. The generally accepted criterion for the definition of a nonclassical field resides in the existence of a positive P-function, i.e., a classical state is one whose P-function is no more singular than a δ-function and is nonnegative definite (e.g., Refs. 23, 98, 202, and 203). This means that the quantum

statistical properties of the nonclassical field cannot be described completely within the framework of a classical probability theory. A detailed discussion of the existence of quasidistributions $\mathscr{W}^{(s)}(\{\alpha_k\})$ for the Raman scattering model under consideration is presented in Section V.B. The Wigner function always exists as a nonsingular function, but may assume negative values, and in this sense is not a classical probability distribution (nevertheless, as was shown by Stenholm,[204] experiments always give a positive Wigner function). The Q-function has the properties of a well-behaved (bounded, nonnegative and infinitely differentiable) classical probability distribution.

Let us write down the relation between two s_1- and s_2-parametrized quasidistributions:

$$\mathscr{W}^{(s_2)}(\{\alpha_k\}, t) = \left(\frac{2}{s_1 - s_2}\right)^M \int \exp\left(-\frac{2}{s_1 - s_2}\sum_k |\alpha_k - \beta_k|^2\right) \times \mathscr{W}^{(s_1)}(\{\beta_k\}, t)\, \mathrm{d}^2\left\{\frac{\beta_k}{\pi}\right\} \tag{26}$$

where $s_2 < s_1$. It is seen that the quasidistribution $\mathscr{W}^{(s_2)}$ is given by the convolution of $\mathscr{W}^{(s_1)}$ with the multidimensional Gaussian distribution. The analogous relation for characteristic functions (20) is simpler and valid for any s_1 and s_2,

$$\mathscr{C}^{(s_2)}(\{\beta_k\}, t) = \mathscr{C}^{(s_1)}(\{\beta_k\}, t) \exp\left(\frac{s_2 - s_1}{2}\sum_k |\beta_k|^2\right) \tag{27}$$

Even in the case when s_1-parametrized QPDs do not exist, the calculation of the expectation values $\langle \hat{a}^{+m}\hat{a}^n\rangle_{(s_1)}$ in s_1 order poses no problem. They can be obtained from the corresponding s_1-parametrized characteristic function $\mathscr{C}^{(s_1)}$ in view of Eq. (25) or, equivalently, from an s_2-parametrized quasidistribution $\mathscr{W}^{(s_2)}$, which does exist, by means of the relation

$$\left\langle \prod_k (\hat{a}_k^+)^{m_k} (\hat{a}_k)^{n_k} \right\rangle_{(s_1)} = \int \sum_k m_k! \left(\frac{s_2 - s_1}{2}\right)^{m_k} \times \alpha_k^{n_k - m_k} L_{m_k}^{n_k - m_k}\left(\frac{2|\alpha_k|^2}{s_1 - s_2}\right) \mathscr{W}^{(s_2)}(\{\alpha_k\})\, \mathrm{d}^2\left\{\frac{\alpha_k}{\pi}\right\} \tag{28}$$

where $L_m^n(x)$ is the generalized Laguerre polynomial. Alternatively, to

obtain the s_1-ordered moments $\langle \hat{a}^{+m}\hat{a}^n\rangle_{(s_1)}$ one can use the generalized P-representation (positive P-representation).[124-128]

IV. PHOTON-COUNTING STATISTICS AND SQUEEZING: DEFINITIONS

To investigate nonclassical phenomena such as sub-Poissonian photon-counting statistics or photon antibunching, one needs to know the diagonal matrix elements in Fock representation of the density matrix $\hat{\rho}(\{\hat{\alpha}_k\})$ only. We start from the probability distribution $p(n)$ of the photon-number n in the k-mode field within a given volume V of space at the time t, defined by

$$p(n) = \sum_{\{n_k\}} \langle \{n_k\}|\hat{\rho}|\{n_k\}\rangle \delta_{n,\Sigma n_k} \tag{29}$$

where $n_k = |\alpha_k|^2$. The s-parametrized quasidistribution $\mathscr{W}^{(s)}(\{\alpha_k\}, t)$ (21) can be readily transformed to the following s-parametrized integrated quasidistribution (intensity distribution) $\mathscr{W}^{(s)}(W, t)$ by means of the δ-function,

$$\mathscr{W}^{(s)}(W,t) = \int \mathscr{W}^{(s)}(\{\alpha_k\},t)\delta\left(\sum_k |\alpha_k|^2 - W\right) \mathrm{d}^2\left\{\frac{\alpha_k}{\pi}\right\} \tag{30}$$

where the variable W can be interpreted as the integrated intensity. The photodetection equation gives a connection between the continuous integrated quasidistribution $\mathscr{W}^{(1)}(W, t)$ and the discrete photon-number distribution first derived by Mandel.[205, 206] This photodetection equation states that the photocount distribution $p(n)$ is the Poisson transform of the integrated quasidistribution $\mathscr{W}^{(1)}(W, t)$. A generalized photodetection equation for $\mathscr{W}^{(s)}(\{\alpha_k\}, t)$ or for $\mathscr{W}^{(s)}(W, t)$ can be written as[23, 207]

$$\begin{aligned} p(n) &= \left(\frac{2}{1+s}\right)^M \left(\frac{s-1}{1+s}\right)^n \int \mathscr{W}^{(s)}(\{\alpha_k\}) \\ &\quad \times \exp\left(-\frac{2}{1+s}\sum_k |\alpha_k|^2\right) L_n^{M-1}\left(\frac{4}{1-s^2}\sum_k |\alpha_k|^2\right) \mathrm{d}^2\left\{\frac{\alpha_k}{\pi}\right\} \\ &= \left(\frac{2}{1+s}\right)^M \left(\frac{s-1}{1+s}\right)^n \int \mathscr{W}^{(s)}(W) \\ &\quad \times \exp\left(-\frac{2W}{1+s}\right) L_n^{M-1}\left(\frac{4W}{1-s^2}\right) \mathrm{d}^2 W \end{aligned} \tag{31}$$

with $L_n^{M-1}(x)$ denoting the generalized Laguerre polynomial. We formally identify the photon-number distribution (29) with the photocount distribution (30). There is some slight difference in their physical interpretation, since the former distribution describes the probability of having n photons in the mode volume V, whereas the latter distribution describes the probability of detecting n photons in the detector volume V_{det}, defined by its parameters (sensitive area, response time, quantum efficiency, etc.). It can be argued, however (e.g., Refs. 208–210), that there is perfect physical equivalence between the photon-number moments obtained from (29) and the photocount-number moments calculated from (31), under the assumption of ideal detectors.

The s-parametrized time-dependent generating function $\langle \exp(-\lambda W)\rangle_{(s)}$, defined by the Fourier transform of the s-parametrized quasidistribution $\mathscr{W}^{(s)}(\{\alpha_k\}, t)$ or characteristic function $\mathscr{C}^{(s)}(\{\beta_k\}, t)$:

$$\begin{aligned}\langle \exp(-\lambda W(t))\rangle_{(s)} &= \int \mathscr{W}^{(s)}(\{\alpha_k\}, t) \exp\left(-\lambda \sum_k |\alpha_k|^2\right) \mathrm{d}^2\left\{\frac{\alpha_k}{\pi}\right\} \\ &= \lambda^{-M} \int \mathscr{C}^{(s)}(\{\beta_k\}, t) \exp\left(-\frac{1}{\lambda} \sum_k |\beta_k|^2\right) \mathrm{d}^2\left\{\frac{\alpha_k}{\pi}\right\}\end{aligned} \tag{32}$$

enables us to calculate the photon-number distribution $p(n, t)$ and the s-ordered photon-number moments $\langle \hat{n}^k \rangle_{(s)}$ in a particularly simple manner:

$$p(n) = \frac{(-1)^n}{n!} \frac{\mathrm{d}^n}{\mathrm{d}\lambda^n} \left(1 + \frac{s-1}{2}\lambda\right)^{-M} \left\langle \exp\left(-\frac{\lambda}{1 + \frac{s-1}{2}\lambda} W\right) \right\rangle_{(s)} \Bigg|_{\lambda = 1} \tag{33}$$

$$\langle W^k \rangle_{(s)} = (-1)^k \frac{\mathrm{d}^k}{\mathrm{d}\lambda^k} \langle \exp(-\lambda W)\rangle_{(s)} \Big|_{\lambda = 0} \tag{34}$$

Eq. (33) takes the simplest form for $s = 1$. Several parameters are widely used in the literature to describe the photon-number statistics, e.g.; the Mandel Q parameter, the Fano factor, or the normalized second-order correlation function. In our analysis we employ the normalized second-order factorial moment of the photon-number operators (or integrated

intensity) (Refs. 23 and 210 and references therein)

$$\gamma_k^{(2)} = \frac{\langle(\Delta\hat{n}_k)^2\rangle_{(1)}}{\langle\hat{n}_k\rangle^2} = \frac{\langle\hat{n}_k^2\rangle_{(1)}}{\langle\hat{n}_k\rangle^2} - 1 = \frac{\langle\hat{n}_k(\hat{n}_k - 1)\rangle}{\langle\hat{n}_k\rangle^2} - 1 \tag{35}$$

and its generalization, the normalized pth order factorial moment of the kth and lth mode (the normalized two-mode cross-correlation function of pth order)

$$\gamma_{kl}^{(p)} = \frac{\langle\hat{n}_{kl}^p\rangle_{(1)}}{\langle\hat{n}_{kl}\rangle^p} - 1 = \frac{\langle\hat{n}_{kl}(\hat{n}_{kl} - 1)\ldots(\hat{n}_{kl} - p + 1)\rangle}{\langle\hat{n}_{kl}\rangle^p} - 1 \tag{36}$$

where $\hat{n}_{kl} = \hat{n}_k + \hat{n}_l$. The higher-order factorial moments (36) by comparison with the second-order moments (35) provide us with further information concerning the photon-number distributions. In view of the fact that $\hat{n}_{kl}$ is the sum of the single-mode photon-number operators, the factorial moment $\gamma_{kl}^{(2)}$ can be written as

$$\gamma_{kl}^{(2)} = \frac{\langle(\Delta\hat{n}_{kl})^2\rangle_{(1)}}{\langle\hat{n}_{kl}\rangle^2} = \frac{\langle(\Delta\hat{n}_k)^2\rangle_{(1)} + \langle(\Delta\hat{n}_l)^2\rangle_{(1)} + 2\langle\Delta\hat{n}_k\,\Delta\hat{n}_l\rangle}{\langle\hat{n}_k\rangle^2 + \langle\hat{n}_l\rangle^2 + 2\langle\hat{n}_k\rangle\langle\hat{n}_l\rangle} \tag{37}$$

The Mandel Q parameter for the mode k is equal to $\gamma_k^{(2)}\langle\hat{n}_k\rangle$, whereas the Fano factor F is $(\gamma_k^{(2)}\langle\hat{n}_k\rangle + 1)$ (the photoefficiency η of the photodetector is assumed to be $\eta = 1$).

Light with photon-number fluctuations smaller than those of the Poisson distribution is called sub-Poissonian (or photon-number squeezed) and is described by a negative value of $\gamma^{(2)}$, both for $\gamma_k^{(2)}$ in the single-mode case and for $\gamma_{kl}^{(2)}$ in the two-mode case. In Section VI we analyze the two-mode model of the Raman effect that comprises the laser (L) and the Stokes mode (S). We show that the sum of photon-number operators in both modes is a constant of motion, which implies that the factorial moments $\gamma_{LS}^{(p)}$ are constant as well. Henceforth we shall be applying another definition to investigate two-mode cross-correlation, referred to as the interbeam degree of second-order coherence, given by (Ref. 30 and references therein)

$$g_{kl}^{(2)} = \frac{\langle\Delta\hat{n}_k\,\Delta\hat{n}_l\rangle}{\langle\hat{n}_k\rangle\langle\hat{n}_l\rangle} = \frac{\langle\hat{n}_k\hat{n}_l\rangle}{\langle\hat{n}_k\rangle\langle\hat{n}_l\rangle} - 1 \tag{38}$$

(Our definition deviates from those of Ref. 30 by the extra term -1.)

To investigate squeezing properties of light we introduce the Hermitian single- and two-mode operators:

$$\hat{X}_k(\theta) = \hat{a}_k \, e^{-i\theta} + \hat{a}_k^+ \, e^{i\theta} \tag{39}$$

$$\hat{X}_{jk}(\theta) = \hat{a}_{jk} \, e^{-i\theta} + \hat{a}_{jk}^+ \, e^{i\theta} = \hat{X}_j(\theta) + \hat{X}_k(\theta) \tag{40}$$

where $\hat{a}_{kl} = \hat{a}_k + \hat{a}_l$. The operator $\hat{X}_k(\hat{X}_{kl})$ for $\theta = 0$ corresponds to the in-phase quadrature component of the kth (kth and lth) mode (modes) of the field, whereas for $\theta = \pi/2$ it corresponds to the out-of-phase component. For brevity, we use the notation $\hat{X}_{k1} = \hat{X}_k(0)$, $\hat{X}_{k2} = \hat{X}_k(\pi/2)$, as well as $\hat{X}_{kl1} = \hat{X}_{kl}(0)$ and $\hat{X}_{kl2} = \hat{X}_{kl}(\pi/2)$. The following commutation rules hold:

$$\left[\hat{X}_{k1}, \hat{X}_{k2}\right] = 2i \tag{41}$$

$$\left[\hat{X}_{kl1}, \hat{X}_{kl2}\right] = 4i \tag{42}$$

Firstly, we shall discuss in brief the single-mode case. The variances of the θ-dependent quadrature (39) are

$$\left\langle \left(\Delta \hat{X}_k(\theta)\right)^2 \right\rangle = 2 \operatorname{Re}\left[e^{-2i\theta} \langle (\Delta \hat{a}_k)^2 \rangle\right] + \left\langle \{\Delta \hat{a}_k^+, \Delta \hat{a}_k\} \right\rangle \tag{43}$$

which obviously give $\hat{X}_{k1}$ and $\hat{X}_{k2}$ in special cases. The Heisenberg uncertainty relation for quadratures,

$$\left\langle \left(\Delta \hat{X}_{k1}\right)^2 \right\rangle \left\langle \left(\Delta \hat{X}_{k2}\right)^2 \right\rangle \geq 1 \tag{44}$$

lays the basis for the definition of "usual" ("standard") squeezing. The state of the field is said to be squeezed if the variance of $\hat{X}_{k1}$ or $\hat{X}_{k2}$ becomes smaller than unity (in general, smaller than the square root of the right side of the uncertainty relation for the quadratures). Equivalently, light whose quantum fluctuations in the one quadrature are smaller than those associated with coherent light (minimizing the uncertainty relation) is called squeezed (in the usual meaning). Since, for a given quantum state, the variance (43) is still dependent on θ, the angle θ can be chosen in a way to minimize (or maximize) the variance. Differentiation with respect to θ leads to the angles θ_+ and θ_- for the maximal and minimal

variances, respectively, given by the relation[211, 212]

$$\exp(2i\theta_{\pm}) = \pm\left(\frac{\langle(\Delta\hat{a}_k)^2\rangle}{\langle(\Delta\hat{a}_k^+)^2\rangle}\right)^{1/2} \tag{45}$$

where the difference between the angles θ_+ and θ_- is $\pi/2$. On inserting (45) into (43) one obtains the extremal variances

$$\begin{aligned}\left\langle\left(\Delta\hat{X}_{k\pm}\right)^2\right\rangle &\equiv \left\langle\left(\Delta\hat{X}_k(\theta_{\pm})\right)^2\right\rangle \\ &= \pm 2\left|\left\langle(\Delta\hat{a}_k)^2\right\rangle\right| + \left\langle\{\Delta\hat{a}_k^+, \Delta\hat{a}_k\}\right\rangle\end{aligned} \tag{46}$$

It is noteworthy that the θ-dependent variance (43) can be expressed in terms of the extremal variances

$$\begin{aligned}\left\langle\left(\Delta\hat{X}_k(\theta)\right)^2\right\rangle = &\left\langle\left(\Delta\hat{X}_{k-}\right)^2\right\rangle\cos^2(\theta - \theta_-) \\ &+ \left\langle\left(\Delta\hat{X}_{k+}\right)^2\right\rangle\sin^2(\theta - \theta_-)\end{aligned} \tag{47}$$

which is the equation for Booth's elliptical lemniscate in polar coordinates.[212] The principal squeezing, introduced by Lukš et al.,[211, 213] occurs if the minimum variance is less than unity:

$$\left\langle\left(\Delta\hat{X}_{k-}\right)^2\right\rangle \leq 1 \tag{48}$$

From (46) it follows that the principal squeezing requires the fulfillment of the condition

$$\langle\Delta\hat{a}_k^+\Delta\hat{a}_k\rangle < \left|\left\langle(\Delta\hat{a}_k)^2\right\rangle\right| \tag{49}$$

whereas the condition for standard squeezing, in view of (43), is

$$\min\left\{\langle\Delta\hat{a}_k^+\Delta\hat{a}_k\rangle \pm \mathrm{Re}\left[\left\langle(\Delta\hat{a}_k)^2\right\rangle\right]\right\} < 0 \tag{50}$$

The mathematically elegant formalism of principal squeezing (in particular other equivalent conditions for principal squeezing) can be formulated using the generalized Heisenberg uncertainty relation (the Schrödinger

uncertainty relation)[211, 214, 215]:

$$\left\langle \left(\Delta \hat{X}_{k1}\right)^2 \right\rangle \left\langle \left(\Delta \hat{X}_{k2}\right)^2 \right\rangle \geq \tfrac{1}{4} \left\langle \left\{ \Delta \hat{X}_{k1}, \Delta \hat{X}_{k2} \right\} \right\rangle^2 + 1 \tag{51}$$

which includes the Wigner covariance (cross-correlation) of the quadratures $\hat{X}_{k1}$ and $\hat{X}_{k2}$ equal to

$$\left\langle \left\{ \Delta \hat{X}_{k1}, \Delta \hat{X}_{k2} \right\} \right\rangle = 4 \operatorname{Im} \left[\left\langle \left(\Delta \hat{a}_k\right)^2 \right\rangle \right] \tag{52}$$

For extremal variances $\langle (\Delta \hat{X}_{k\pm})^2 \rangle$ the generalized Heisenberg relation reduces to the standard uncertainty relation.

The generalization of the above definitions for the two-mode case is straightforward. By virtue of the commutator (42), twice as great as for the single-mode case (41), the standard and principal squeezing can be defined, respectively, as

$$\min \left\{ \left\langle \left(\Delta \hat{X}_{kl1}\right)^2 \right\rangle, \left\langle \left(\Delta \hat{X}_{kl2}\right)^2 \right\rangle \right\} \leq 2 \tag{53}$$

$$\left\langle \left(\Delta \hat{X}_{kl-}\right)^2 \right\rangle \leq 2 \tag{54}$$

We express the two-mode variances and the Wigner covariances in terms of the single-mode moments:

$$\left\langle \left(\Delta \hat{X}_{kl}\right)^2 \right\rangle = \left\langle \left(\Delta \hat{X}_{k}\right)^2 \right\rangle + \left\langle \left(\Delta \hat{X}_{l}\right)^2 \right\rangle + 2 \left\langle \Delta \hat{X}_k \Delta \hat{X}_l \right\rangle \tag{55}$$

$$\begin{aligned} \left\langle \left\{ \Delta \hat{X}_{kl1}, \Delta \hat{X}_{kl2} \right\} \right\rangle = {} & \left\langle \left\{ \Delta \hat{X}_{k1}, \Delta \hat{X}_{k2} \right\} \right\rangle + \left\langle \left\{ \Delta \hat{X}_{l1}, \Delta \hat{X}_{l2} \right\} \right\rangle \\ & + 2 \left\langle \Delta \hat{X}_{k1} \Delta \hat{X}_{l2} \right\rangle + 2 \left\langle \Delta \hat{X}_{k2} \Delta \hat{X}_{l1} \right\rangle \end{aligned} \tag{56}$$

where $\hat{X}_{kl}$ stands for $\hat{X}_{kl}(\theta)$ (in particular the quadratures). Relations such as (55) and (56) for the quadratures hold for the two-mode creation and annihilation operators $\hat{a}^{\pm}_{kl}$. The moments $\langle (\hat{X}_k)^2 \rangle$ and $\langle \{\Delta \hat{X}_{k1}, \Delta \hat{X}_{k2}\} \rangle$ are given by (43) and (52). The remaining cross-correlations have the following form in terms of the annihilation and creation operators:

$$\begin{aligned} \langle \Delta \hat{X}_{k1} \Delta \hat{X}_{l1} \rangle &= 2 \operatorname{Re} \left[\langle \Delta \hat{a}_k \Delta \hat{a}_l \rangle + \langle \Delta \hat{a}_k^+ \Delta \hat{a}_l \rangle \right] \\ \langle \Delta \hat{X}_{k2} \Delta \hat{X}_{l2} \rangle &= 2 \operatorname{Re} \left[-\langle \Delta \hat{a}_k \Delta \hat{a}_l \rangle + \langle \Delta \hat{a}_k^+ \Delta \hat{a}_l \rangle \right] \\ \langle \Delta \hat{X}_{k1} \Delta \hat{X}_{l2} \rangle &= 2 \operatorname{Im} \left[\langle \Delta \hat{a}_k \Delta \hat{a}_l \rangle + \langle \Delta \hat{a}_k^+ \Delta \hat{a}_l \rangle \right] \\ \langle \Delta \hat{X}_{k2} \Delta \hat{X}_{l1} \rangle &= 2 \operatorname{Im} \left[\langle \Delta \hat{a}_k \Delta \hat{a}_l \rangle - \langle \Delta \hat{a}_k^+ \Delta \hat{a}_l \rangle \right] \end{aligned} \tag{57}$$

Substituting Eqs. (43), (52), and (57) into (55) and (56) one obtains explicit dependencies of the two-mode quadrature moments on the annihilation operators.[213]

Alternatively, the single-mode moments (43), (46), and (52) and the conditions (49) and (50) for single-mode squeezing can be generalized to a two-mode case by simple replacement of $\hat{a}_k$, $\hat{X}_k(\theta)$ by $\hat{a}_{kl}$, $\hat{X}_{kl}(\theta)$, showing complete analogy between the single- and two-mode descriptions. In particular, the two-mode extremal variances are

$$\left\langle \left(\Delta \hat{X}_{kl\pm}\right)^2 \right\rangle = \pm 2 \left| \left\langle \left(\Delta \hat{a}_{kl}\right)^2 \right\rangle \right| + \left\langle \left\{ \Delta \hat{a}_{kl}^{+}, \Delta \hat{a}_{kl} \right\} \right\rangle \tag{58}$$

by analogy to (46).

V. FOKKER-PLANCK EQUATION

A. Raman Scattering Including Pump Depletion

The master equation (ME) is the quantum equation of motion for operators and hence it is possible to solve it directly only for a small class of models. As an example we cite the Raman-coupled model of Knight[133] and its generalizations (Ref. 137 and references therein). Usually the quantum master equation is converted to a classical differential equation. Then, standard methods of mathematical analysis can be applied. In this section we present one of the most popular methods: transformation to a generalized Fokker-Planck equation (FPE) or equivalently to an equation of motion for characteristic functions. This method is extensively studied in a number of textbooks[23, 183, 184, 188] and consists of performing s-ordering of the field operators in the ME (18) and then applying the quantum–classical number correspondence of coherent-state technique. The rules for the transformation of the ME into Fokker-Planck equations for the s-parametrized quasidistribution $\mathscr{W}^{(s)}(\alpha, \alpha^*, \bar{A})$ are the following (e.g., Refs. 216 and 217):

$$\begin{aligned} \begin{Bmatrix} \hat{A}\hat{a} \\ \hat{a}\hat{A} \end{Bmatrix} &\mapsto \left(\alpha - \frac{s \pm 1}{2} \frac{\partial}{\partial \alpha^*} \right) \mathscr{W}^{(s)}(\alpha, \alpha^*, \bar{A}) \\ \begin{Bmatrix} \hat{a}^{+}\hat{A} \\ \hat{A}\hat{a}^{+} \end{Bmatrix} &\mapsto \left(\alpha^* - \frac{s \pm 1}{2} \frac{\partial}{\partial \alpha} \right) \mathscr{W}^{(s)}(\alpha, \alpha^*, \bar{A}) \end{aligned} \tag{59}$$

where $\hat{A}$ is an arbitrary operator; in particular, $\hat{A}$ can be the density matrix $\hat{\rho}$; $\bar{A}$ is the classical function associated with the operator $\hat{A}$; the

parameter s takes arbitrary values in the range $\langle -1, 1\rangle$. If necessary, these rules can be applied repeatedly. Similarly, we list the rules of transformation of the master equation (18) to the equation of motion for the s-parametrized characteristic function $\mathscr{C}^{(s)}(\beta, \beta^*, \bar{A})$ (e.g., Ref. 216):

$$\begin{aligned}
\begin{Bmatrix} \hat{A}\hat{a} \\ \hat{a}\hat{A} \end{Bmatrix} &\mapsto \left(-\frac{\partial}{\partial \beta^*} + \frac{s \pm 1}{2}\beta \right) \mathscr{C}^{(s)}(\beta, \beta^*, \bar{A}) \\
\begin{Bmatrix} \hat{a}^+\hat{A} \\ \hat{A}\hat{a}^+ \end{Bmatrix} &\mapsto \left(\frac{\partial}{\partial \beta} - \frac{s \pm 1}{2}\beta^* \right) \mathscr{C}^{(s)}(\beta, \beta^*, \bar{A})
\end{aligned} \tag{60}$$

Applying repeatedly the rule (59) of transformation to the master equation (18) and after some lengthy algebra we finally arrive at the generalized Fokker-Planck equation for the s-parametrized quasidistribution $\mathscr{W}^{(s)} \equiv \mathscr{W}^{(s)}(\alpha_L, \alpha_S, \alpha_A, t)$:

$$\begin{aligned}
\frac{\partial}{\partial t}\mathscr{W}^{(s)} = \frac{1}{2}\gamma_S \Bigg\{ \Bigg[& -\frac{\partial}{\partial \alpha_L}\alpha_L \frac{\partial}{\partial \alpha_S}\alpha_S \\
& + \left(|\alpha_S|^2 + \frac{1+s}{2} \right) \frac{\partial}{\partial \alpha_L}\alpha_L - \left(|\alpha_L|^2 - \frac{1-s}{2} \right) \frac{\partial}{\partial \alpha_S}\alpha_S \\
& + \frac{1-s^2}{4} \left(\frac{\partial}{\partial \alpha_L}\alpha_L \frac{\partial}{\partial \alpha_S}\frac{\partial}{\partial \alpha_S^*} - \frac{\partial}{\partial \alpha_L}\frac{\partial}{\partial \alpha_L^*}\frac{\partial}{\partial \alpha_S}\alpha_S \right) + \text{c.c.} \Bigg] \\
& + \left[(1-s)|\alpha_S|^2 + \frac{1-s^2}{2} \right] \frac{\partial}{\partial \alpha_L}\frac{\partial}{\partial \alpha_L^*} \\
& + \left[(1+s)|\alpha_L|^2 - \frac{1-s^2}{2} \right] \frac{\partial}{\partial \alpha_S}\frac{\partial}{\partial \alpha_S^*} \Bigg\} \mathscr{W}^{(s)} \\
+ \frac{1}{2}\gamma_A \Bigg\{ \Bigg[& -\frac{\partial}{\partial \alpha_L}\alpha_L \frac{\partial}{\partial \alpha_A}\alpha_A \\
& - \left(|\alpha_A|^2 - \frac{1-s}{2} \right) \frac{\partial}{\partial \alpha_L}\alpha_L + \left(|\alpha_L|^2 + \frac{1+s}{2} \right) \frac{\partial}{\partial \alpha_A}\alpha_A \\
& - \frac{1-s^2}{4} \left(\frac{\partial}{\partial \alpha_L}\alpha_L \frac{\partial}{\partial \alpha_A}\frac{\partial}{\partial \alpha_A^*} - \frac{\partial}{\partial \alpha_L}\frac{\partial}{\partial \alpha_L^*}\frac{\partial}{\partial \alpha_A}\alpha_A \right) + \text{c.c.} \Bigg]
\end{aligned}$$

$$+\left[(1+s)|\alpha_A|^2-\frac{1-s^2}{2}\right]\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_L^*}$$

$$+\left[(1-s)|\alpha_L|^2+\frac{1-s^2}{2}\right]\frac{\partial}{\partial\alpha_A}\frac{\partial}{\partial\alpha_A^*}\Bigg\}\mathscr{W}^{(s)}$$

$$+\left\{\frac{1}{2}\gamma_{SA}\exp(-2i\Delta\Omega\,\Delta t)\left[-\alpha_L^2\left(\alpha_A^*\frac{\partial}{\partial\alpha_S}+\frac{\partial}{\partial\alpha_S}\frac{\partial}{\partial\alpha_A}-\alpha_S^*\frac{\partial}{\partial\alpha_A}\right)\right.\right.$$

$$+\alpha_L\left((1+s)\alpha_A^*\frac{\partial}{\partial\alpha_S}+(1-s)\alpha_S^*\frac{\partial}{\partial\alpha_A}\right)\frac{\partial}{\partial\alpha_L^*}$$

$$\left.\left.+\left(\frac{1-s^2}{4}\alpha_A^*\frac{\partial}{\partial\alpha_S}-\alpha_S^*\alpha_A^*-\frac{1-s^2}{4}\alpha_S^*\frac{\partial}{\partial\alpha_A}\right)\frac{\partial^2}{\partial\alpha_L^{*2}}\right]+\text{c.c.}\right\}\mathscr{W}^{(s)}$$

$$+\gamma_S\langle\hat{n}_V\rangle\left\{\left(\frac{1}{2}\frac{\partial}{\partial\alpha_L}\alpha_L-\frac{\partial}{\partial\alpha_L}\alpha_L\frac{\partial}{\partial\alpha_S}\alpha_S+\frac{1}{2}\frac{\partial}{\partial\alpha_S}\alpha_S+\text{c.c.}\right)\right.$$

$$\left.+|\alpha_S|^2\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_L^*}+|\alpha_L|^2\frac{\partial}{\partial\alpha_S}\frac{\partial}{\partial\alpha_S^*}\right\}\mathscr{W}^{(s)}$$

$$+\gamma_A\langle\hat{n}_V\rangle\left\{\left(\frac{1}{2}\frac{\partial}{\partial\alpha_L}\alpha_L-\frac{\partial}{\partial\alpha_L}\alpha_L\frac{\partial}{\partial\alpha_A}\alpha_A+\frac{1}{2}\frac{\partial}{\partial\alpha_A}\alpha_A+\text{c.c.}\right)\right.$$

$$\left.+|\alpha_A|^2\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_L^*}+|\alpha_L|^2\frac{\partial}{\partial\alpha_A}\frac{\partial}{\partial\alpha_A^*}\right\}\mathscr{W}^{(s)}$$

$$-\left\{\gamma_{AS}\langle\hat{n}_V\rangle\exp(2i\Delta\Omega\,\Delta t)\left[\alpha_S^*\alpha_A^*\frac{\partial^2}{\partial\alpha_L^{*2}}+\alpha_L^2\frac{\partial}{\partial\alpha_S}\frac{\partial}{\partial\alpha_A}\right.\right.$$

$$\left.\left.-\alpha_L\left(\alpha_A^*\frac{\partial^2}{\partial\alpha_S}+\alpha_S^*\frac{\partial^2}{\partial\alpha_A}\right)\frac{\partial}{\partial\alpha_L^*}\right]+\text{c.c.}\right\}\mathscr{W}^{(s)} \tag{61}$$

which is a generalization of our former relation for $\gamma_A, \gamma_{SA} \neq 0$ and arbitrary parameter s (Ref. 218). For brevity, we refer to the generalized

Fokker-Planck equation[23] simply as the Fokker-Planck equation (FPE). The physical interpretation of (61) can be given in the same manner as the interpretation of the appropriate terms in the ME (18) given in Section II. The FPE (61) exhibits a highly complicated structure. Nonetheless, the equations of motion for the mean values $\langle \alpha_k \rangle, \langle \alpha_k \alpha_l \rangle, \langle \alpha_k^* \alpha_l \rangle$, (with $k, l = L, S, A$) can be calculated. In particular, we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\left(\langle \hat{n}_L(t) \rangle + \langle \hat{n}_S(t) \rangle + \langle \hat{n}_A(t) \rangle\right) = 0 \tag{62}$$

with $\langle \hat{n}_k(t) \rangle = \langle \alpha_k^* \alpha_k \rangle$. Eq. (62) states that the total mean number of photons (in all radiation modes) is a constant of motion.

The FPE (61) contains terms of the form

$$\frac{\partial}{\partial \alpha_i} \alpha_j \alpha_k \alpha_l \mathscr{W}^{(s)} \qquad \frac{\partial}{\partial \alpha_i} \frac{\partial}{\partial \alpha_j} \alpha_k \alpha_l \mathscr{W}^{(s)} \qquad \frac{\partial}{\partial \alpha_i} \frac{\partial}{\partial \alpha_j} \frac{\partial}{\partial \alpha_k} \alpha_l \mathscr{W}^{(s)}$$

where $\alpha_i, \alpha_j, \alpha_k, \alpha_l = \alpha_L, \alpha_L^*, \alpha_S, \alpha_S^*, \alpha_A, \alpha_A^*$. It is seen that most components of the drift vector are nonlinear to the third order in α, and most components of the diffusion matrix are nonlinear up to the second order. It is particularly difficult to solve a differential equation with such nonlinear diffusion and drift coefficients. Besides, the FPE (61) for $\mathscr{W}^{(s)} \equiv \mathscr{W}^{(s)}(\alpha_L, \alpha_S, \alpha_A, t)$ with the parameter $s \neq \pm 1$ contains third-order derivatives in the terms

$$\frac{\partial}{\partial \alpha_i} \frac{\partial}{\partial \alpha_j} \frac{\partial}{\partial \alpha_k} \alpha_l \mathscr{W}^{(s)}$$

This could be expected since in many models,[23, 19] for instance in the anharmonic oscillator model (for references see Ref. 94), there occur third-order derivatives in the FPEs for the Wigner function ($s = 0$).

The corresponding equation of motion for the s-parametrized characteristic function $\mathscr{C}^{(s)}(\beta_L, \beta_S, \beta_A, t)$ can be obtained either from the ME (18) by performing the transformation (60), or from the FPE (61) by means of the Fourier transformation (22) with respect to the variables $\alpha_L, \alpha_S, \alpha_A$.

Finally, we arrive at the equation of motion for $\mathscr{E}^{(s)} \equiv \mathscr{E}^{(s)}(\beta_L, \beta_S, \beta_A, t)$:

$$\begin{aligned}
\frac{\partial}{\partial t}\mathscr{E}^{(s)} = \frac{1}{2}\gamma_S \Bigg\{ \Bigg[& -\beta_L\beta_S \frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_S} + \beta_L\left(-\frac{1+s}{2} + \frac{\partial}{\partial\beta_S}\frac{\partial}{\partial\beta_S^*}\right)\frac{\partial}{\partial\beta_L} \\
& -\beta_S\left(\frac{1-s}{2} + \frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_L^*}\right)\frac{\partial}{\partial\beta_S} \\
& + \frac{1-s^2}{4}\left(|\beta_S|^2\beta_L\frac{\partial}{\partial\beta_L} - |\beta_L|^2\beta_S\frac{\partial}{\partial\beta_S}\right) + \text{c.c.}\Bigg] \\
& + |\beta_L|^2\left[-\frac{1-s^2}{2} + (1-s)\frac{\partial}{\partial\beta_S}\frac{\partial}{\partial\beta_S^*}\right] \\
& + |\beta_S|^2\left[\frac{1-s^2}{2} + (1+s)\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_L^*}\right]\Bigg\}\mathscr{E}^{(s)} \\
+ \frac{1}{2}\gamma_A \Bigg\{ \Bigg[& -\beta_L\beta_A\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_A} - \beta_L\left(\frac{1-s}{2} + \frac{\partial}{\partial\beta_A}\frac{\partial}{\partial\beta_A^*}\right)\frac{\partial}{\partial\beta_L} \\
& + \beta_A\left(-\frac{1+s}{2} + \frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_L^*}\right)\frac{\partial}{\partial\beta_A} \\
& + \frac{1-s^2}{4}\left(-|\beta_A|^2\beta_L\frac{\partial}{\partial\beta_L} + |\beta_L|^2\beta_A\frac{\partial}{\partial\beta_A}\right) + \text{c.c.}\Bigg] \\
& + |\beta_L|^2\left[\frac{1-s^2}{2} + (1+s)\frac{\partial}{\partial\beta_A}\frac{\partial}{\partial\beta_A^*}\right] \\
& + |\beta_A|^2\left[-\frac{1-s^2}{2} + (1-s)\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_L^*}\right]\Bigg\}\mathscr{E}^{(s)} \\
+ \Bigg\{ \frac{1}{2}\gamma_{SA}\exp(-2i\Delta\Omega\,\Delta t) \Bigg[& \left(\beta_A\frac{\partial}{\partial\beta_S^*} - \beta_S\beta_A - \beta_S\frac{\partial}{\partial\beta_A^*}\right)\frac{\partial^2}{\partial\beta_L^2} \\
& + \beta_L^*\left((1-s)\beta_A\frac{\partial}{\partial\beta_S^*} + (1+s)\beta_S\frac{\partial}{\partial\beta_A^*}\right)\frac{\partial}{\partial\beta_L} \\
& - \beta_L^{*2}\left(\frac{1-s^2}{4}\beta_A\frac{\partial}{\partial\beta_S^*} + \frac{\partial}{\partial\beta_S^*}\frac{\partial}{\partial\beta_A^*} - \frac{1-s^2}{4}\beta_S\frac{\partial}{\partial\beta_A^*}\right)\Bigg] + \text{c.c.}\Bigg\}\mathscr{E}^{(}
\end{aligned}$$

$$
\begin{aligned}
&+\gamma_S\langle\hat{n}_V\rangle\left\{-\left(\frac{1}{2}\beta_L\frac{\partial}{\partial\beta_L}+\beta_L\beta_S\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_S}+\frac{1}{2}\beta_S\frac{\partial}{\partial\beta_S}+\text{c.c.}\right)\right.\\
&\left.+|\beta_S|^2\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_L^*}+|\beta_L|^2\frac{\partial}{\partial\beta_S}\frac{\partial}{\partial\beta_S^*}\right\}\mathscr{E}^{(s)}\\
&+\gamma_A\langle\hat{n}_V\rangle\left\{-\left(\frac{1}{2}\beta_L\frac{\partial}{\partial\beta_L}+\beta_L\beta_A\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_A}+\frac{1}{2}\beta_A\frac{\partial}{\partial\beta_A}+\text{c.c.}\right)\right.\\
&\left.+|\beta_A|^2\frac{\partial}{\partial\beta_L}\frac{\partial}{\partial\beta_L^*}+|\beta_L|^2\frac{\partial}{\partial\beta_A}\frac{\partial}{\partial\beta_A^*}\right\}\mathscr{E}^{(s)}\\
&-\left\{\frac{1}{2}\gamma_{AS}\langle\hat{n}_V\rangle\exp(2i\Delta\Omega\,\Delta t)\left[\beta_S\beta_A\frac{\partial^2}{\partial\beta_L^2}+\beta_L^{*2}\frac{\partial}{\partial\beta_S^*}\frac{\partial^2}{\partial\beta_A^*}\right.\right.\\
&\left.\left.-\beta_L^*\left(\beta_S\frac{\partial}{\partial\beta_A^*}+\beta_A\frac{\partial}{\partial\beta_S^*}\right)\frac{\partial}{\partial\beta_L}\right]+\text{c.c.}\right\}\mathscr{E}^{(s)}
\end{aligned}
\tag{63}
$$

Here, we come upon similar difficulties in the way of obtaining an analytical solution of (63) as in the FPE case (61), inherent in the nonlinearity of the coefficients of the terms with first- and second-order derivatives as well as the presence of terms with third-order derivatives. Nevertheless, in contradistinction to $\mathscr{W}^{(s)}$, the existence of a solution for $\mathscr{E}^{(s_1)}$ implies, in view of the property (27), the existence of a solution for any other parameter s_2.

In view of the particularly complicated structure of Eqs. (61) and (63) or equivalent equations of motion derived within the completely quantum model of scattering into both the Stokes and anti-Stokes modes, it would seem that a solution in exact closed form cannot be obtained.[23] It is necessary to apply further restrictions or approximations in the model to achieve an analytical solution of (61). In Section VI.A.2 we present a strict analytical solution of the two-mode ME (including pump depletion) in terms of Fock states by applying the Laplace transform. In Section V.B we present solutions of two-mode linearized FPEs for $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ and solutions of equivalent equations of motion for $\mathscr{E}^{(s)}(\beta_S, \beta_A, t)$ in the Raman scattering model under parametric approximation. In Appendix A we give the solution of a linearized form of the three-mode FPE (61) for $\mathscr{W}^{(-1)}(\alpha_L, \alpha_S, \alpha_A, t)$ properly describing the evolution of the radiation fields valid only on the assumption of small fluctuations of the fields around their mean values. There, we restrict our considerations to the

Q-function ($s = -1$) to avoid problems of the existence of the quasidistribution $\mathscr{W}^{(s)}(\alpha_L, \alpha_S, \alpha_A, t)$ (particularly important in the case of s close or equal to 1) and to simplify the third-order FPE (61) to second-order, which takes place for $s = \pm 1$. Within a similar model of Raman scattering from a single-phonon mode, Szlachetka et al.[58-61] (see also Ref. 27) and Tänzler and Schütte[63] have solved the equations of motion in the short-time approximation up to the second power in time. Within the latter (single phonon mode) model Peřina and Křepelka[67, 68] have obtained approximate solutions using the approximation of small fluctuations around a stationary solution.

B. Raman Scattering Without Pump Depletion

Here, to find a solution of the ME (18) we apply the parametric approximation, so no allowance for pump depletion is included. The trilinear Hamiltonians $\hat{H}_A, \hat{H}_S$ (2) can be reduced to bilinear functions as a result of the replacement of the annihilation operator $\hat{a}_L$, representing the quantum pump field, by the classical complex amplitude of the pump field, e_L. This approximation effectively linearizes our model of Raman scattering. Then, the Fokker-Planck equation for the two-mode s-parametrized quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ takes the form

$$\begin{aligned}
\frac{\partial}{\partial t}\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t) = \Bigg\{ & -\left[\left(\frac{\kappa_S}{2} + i\Delta\Omega\right)\frac{\partial}{\partial \alpha_S}\alpha_S + \text{c.c.}\right] \\
& + \left[\left(\frac{\kappa_A}{2} - i\Delta\Omega\right)\frac{\partial}{\partial \alpha_A}\alpha_A + \text{c.c.}\right] \\
& - \left[\frac{\kappa_{SA}}{2}\left(\alpha_A^*\frac{\partial}{\partial \alpha_S} - \alpha_S^*\frac{\partial}{\partial \alpha_A}\right) + \text{c.c.}\right] \\
& + \kappa_S\left(\langle \hat{n}_V \rangle + \frac{s+1}{2}\right)\frac{\partial^2}{\partial \alpha_S \partial \alpha_S^*} \\
& + \kappa_A\left(\langle \hat{n}_V \rangle + \frac{1-s}{2}\right)\frac{\partial^2}{\partial \alpha_A \partial \alpha_A^*} \\
& - \left[\kappa_{SA}\left(\langle \hat{n}_V \rangle + \frac{1}{2}\right)\frac{\partial^2}{\partial \alpha_S \partial \alpha_A} + \text{c.c.}\right]\Bigg\} \\
& \times \mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)
\end{aligned} \tag{64}$$

on applying the rules (59) to the ME (18) with complex classical amplitude e_L instead of the annihilation operator $\hat{a}_L$ and transforming the variables $\alpha_k \to \exp(-i\Delta\Omega\Delta t)\alpha_k$ and $\beta_k \to \exp(-i\Delta\Omega\Delta t)\beta_k$ with the frequency

mismatch $\Delta\Omega$ defined by (12). For brevity, we have incorporated the complex amplitude e_L into the coupling constants $\kappa_S = \gamma_S|e_L|^2$, $\kappa_A = \gamma_A|e_L|^2$, and $\kappa_{SA} = \kappa_{AS}^* = \gamma_{SA}e_L^2$. Equation (64) is a generalization for any parameter s ($s \in \langle -1, 1\rangle$) of the FPE given by Walls for the P-function ($s = 1$)[122] and by Peřina for the P- and Q-functions ($s = \pm 1$).[53, 54] If we consider production of the Stokes radiation only, neglecting anti-Stokes scattering, then Eq. (64) reduces to the s-parametrized FPE obtained by Peřinova et al.[189]

We can interpret the FPE (64) in the same manner as the ME (18). The first term in (64) describes the amplification of the Stokes beam, whereas the second term describes the attenuation of the anti-Stokes beam; the third term shows the coupling between the Stokes and anti-Stokes fields; the remaining three terms account for the noise diffusion from the "noisy" (for nonzero temperature) reservoir into the system. Contrary to the former equations of motion (18), (61), and (60), we lose all information about the depletion of the laser field. It is seen that the FPE (64) for any quasidistribution $\mathscr{W}^{(s)}$, even if related to the field ordering $s \neq \pm 1$, does not contain third-order derivatives, contrary to the FPE (61) without parametric approximation. Let us note that (64) describes an Ornstein-Uhlenbeck process[191] since the components of the drift vector are linear and those of the diffusion matrix are constant. Various methods have been developed for solving the equations of motion for Ornstein-Uhlenbeck processes.[23, 188] For instance, expressing the quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ by its Fourier transform (22) with respect to the variables α_S, α_A, we obtain the following first-order differential equation for the Fourier transform, i.e., for the characteristic function $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$:

$$
\begin{aligned}
\frac{\partial}{\partial t}\mathscr{C}^{(s)}(\beta_S, \beta_A, t) = \Bigg\{ & \left[\left(\frac{\kappa_S}{2} - i\Delta\Omega\right)\beta_S\frac{\partial}{\partial\beta_S} + \text{c.c.}\right] \\
& - \left[\left(\frac{\kappa_A}{2} + i\Delta\Omega\right)\beta_A\frac{\partial}{\partial\beta_A} + \text{c.c.}\right] \\
& + \left[\frac{\kappa_{SA}}{2}\left(\beta_A^*\frac{\partial}{\partial\beta_S} - \beta_S^*\frac{\partial}{\partial\beta_A}\right) + \text{c.c.}\right] \\
& - \kappa_S\left(\langle\hat{n}_V\rangle + \frac{s+1}{2}\right)|\beta_S|^2 \\
& - \kappa_A\left(\langle\hat{n}_V\rangle + \frac{1-s}{2}\right)|\beta_A|^2 \\
& - \left[\kappa_{AS}\left(\langle\hat{n}_V\rangle + \frac{1}{2}\right)\beta_S\beta_A + \text{c.c.}\right]\Bigg\}\mathscr{C}^{(s)}(\beta_S, \beta_A, t)
\end{aligned}
\tag{65}
$$

Again, to obtain (22), one might use the rules (60) applied to (18) with $\hat{a}_L \to e_L$. Our further results presented in this section are mainly based on very extensive studies carried out by Peřina,[53, 54] Peřinová and Peřina,[48] and Kárská and Peřina[56] (see also Ref. 23 and references therein). However, their solutions of the equations of motion for the Raman effect under parametric approximation hold only for quasidistributions $\mathscr{W}^{(1)}(\alpha_S, \alpha_A, t)$ or $\mathscr{W}^{(-1)}(\alpha_S, \alpha_A, t)$ and characteristic functions $\mathscr{C}^{(\pm 1)}(\beta_S, \beta_A, t)$ related to normal and/or antinormal ordering of the field operators. We generalize their results to functions related to s-ordering of the field operators, i.e., to an s-parametrized quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ and s-parametrized characteristic function $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$. Let us use, after Ref. 23, the following simplified notation for functions characterizing the quantum noise, i.e., the Wigner covariances and variances as well as the mean values of the annihilation operators $\hat{a}_S$ and $\hat{a}_A$:

$$
\begin{aligned}
B_k^{(s)}(t) &= \frac{1}{2}\langle\{\Delta\hat{a}_k^+(t), \Delta\hat{a}_k(t)\}\rangle - \frac{s}{2} \\
D_{kl}(t) &= D_{lk}(t) = \frac{1}{2}\langle\{\Delta\hat{a}_k(t), \Delta\hat{a}_l(t)\}\rangle \\
\overline{D}_{kl}(t) &= \overline{D}_{lk}^*(t) = -\frac{1}{2}\langle\{\Delta\hat{a}_k^+(t), \Delta\hat{a}_l(t)\}\rangle \\
C_k(t) &= \langle(\Delta\hat{a}_k(t))^2\rangle \\
\xi_k(t) &= \langle\hat{a}_k(t)\rangle
\end{aligned}
\tag{66}
$$

where $k = S, A$ and $\{\ldots, \ldots\}$ is an anticommutator. Assuming the initial condition that the Stokes and anti-Stokes fields are stochastically independent, the solution of (65) for the s-parametrized characteristic function exists for any parameter s and is equal to

$$
\begin{aligned}
\mathscr{C}^{(s)}(\beta_S, \beta_A, t) = \exp\Bigg\{ \sum_{k=S,A} \Big[& -B_k^{(s)}(t)|\beta_k|^2 \\
& + \left(\tfrac{1}{2}C_k^*(t)\beta_k^2 + \text{c.c.}\right) + \left(\beta_k\xi_k^*(t) - \text{c.c.}\right)\Big] \\
& + \left[D_{SA}(t)\beta_S^*\beta_A^* + \overline{D}_{SA}(t)\beta_S\beta_A^* + \text{c.c.}\right]\Bigg\}
\end{aligned}
\tag{67}
$$

where

$$
\begin{aligned}
B_S^{(s)}(t) &= \left(B_S^{(s)} + \langle \hat{n}_V \rangle + \frac{1+s}{2}\right)|U_S(t)|^2 \\
&\quad + \left(B_A^{(s)} - \langle \hat{n}_V \rangle - \frac{1-s}{2}\right)|V_S(t)|^2 - \langle \hat{n}_V \rangle - \frac{1+s}{2} \\
B_A^{(s)}(t) &= \left(B_A^{(s)} - \langle \hat{n}_V \rangle - \frac{1-s}{2}\right)|U_A(t)|^2 \\
&\quad + \left(B_S^{(s)} + \langle \hat{n}_V \rangle + \frac{1+s}{2}\right)|V_A(t)|^2 + \langle \hat{n}_V \rangle + \frac{1-s}{2} \\
D_{SA}(t) &= \left(B_S^{(s)} + \langle \hat{n}_V \rangle + \frac{1+s}{2}\right)U_S(t)V_A(t) \\
&\quad + \left(B_A^{(s)} - \langle \hat{n}_V \rangle - \frac{1-s}{2}\right)V_S(t)U_A(t) \\
\overline{D}_{SA}(t) &= C_S U_S(t)V_A^*(t) + C_A^* U_A^*(t)V_S(t) \\
C_S(t) &= C_S U_S^2(t) + C_A^* V_S^2(t) \\
C_A(t) &= C_A U_A^2(t) + C_S^* V_A^2(t) \\
\xi_S(t) &= U_S(t)\xi_S + V_S(t)\xi_A^* \\
\xi_A(t) &= U_A(t)\xi_A + V_A(t)\xi_S^*
\end{aligned}
\tag{68}
$$

The solution (67) for $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$ with any parameter s from $\langle -1, 1\rangle$ is, in view of the property (27), a straightforward generalization of the solutions given by Kárská and Peřina[56] (see also Ref. 23) for $\mathscr{C}^{(\pm 1)}(\beta_S, \beta_A, t)$ related with normal or antinormal field operator ordering. Setting initial values $C_A = C_S = 0$, which implies that $\overline{D}_{SA}(t) = C_A(t) = C_S(t) = 0$ for any time t, the solution (67) reduces to that of Peřina.[53] The time-dependent functions $U_k(t)$, $V_k(t)$ $(k = S, A)$ appearing in (68) can be expressed as

$$
\begin{aligned}
V_S(t) &= \frac{\kappa_{SA}}{2} Q_1 \\
V_A(t) &= -\frac{\kappa_{SA}}{2} Q_1^* \\
U_S(t) &= Q_2 + \left(\frac{\kappa_A}{2} + \mathrm{i}\,\Delta\Omega\right)Q_1 \\
U_A(t) &= Q_2^* - \left(\frac{\kappa_S}{2} - \mathrm{i}\,\Delta\Omega\right)Q_1^*
\end{aligned}
\tag{69}
$$

in terms of the auxiliary functions

$$Q_1 = \frac{\exp(P_1 \Delta t) - \exp(P_2 \Delta t)}{P_1 - P_2}$$
$$Q_2 = \frac{\partial Q_1}{\partial t} = \frac{P_1 \exp(P_1 \Delta t) - P_2 \exp(P_2 \Delta t)}{P_1 - P_2} \tag{70}$$

$$P_{1,2} = \frac{1}{2}\left\{\frac{\kappa_S - \kappa_A}{2} \pm \left[\left(\frac{\kappa_S - \kappa_A}{2}\right)^2 - 4\left((\Delta\Omega)^2 - \mathrm{i}\frac{\kappa_S + \kappa_A}{2}\Delta\Omega\right)\right]^{1/2}\right\} \tag{71}$$

It is seen that for the initial moment of time t_0 the functions $V_k(t_0)$ vanish and the $U_k(t_0)$ are equal to unity, so that the initial Wigner covariances $D_{k,l}$, and $\overline{D}_{k,l}$ $(k, l = S, A)$ also vanish as a result of the initial condition of zero stochastical correlation between the scattered modes. Let us note that on the assumption of the frequency resonant condition $\Delta\Omega = 0$, the functions (69)–(71) simplify considerably[53] since $P_1 = \frac{1}{2}(\kappa_S - \kappa_A)$ and $P_2 = 0$. This leads, in particular, to the relations

$$V_S(t) + V_A(t) = 0,$$
$$U_S(t) + U_A(t) = 1 + \exp\left(\frac{\kappa_S - \kappa_A}{2}\Delta t\right) \tag{72}$$

To obtain the solution of the FPE (64) we perform the Fourier transform (21) of $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$, which leads to the s-parametrized quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ in the form

$$\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t) = \frac{1}{L^{(s)}} \exp\left\{(L^{(s)})^{-2}\left[-E_1|\alpha_S - \xi_S(t)|^2 - E_2|\alpha_A - \xi_A(t)|^2 + \frac{1}{2}E_3(\alpha_S^* - \xi_S^*(t))^2 + \frac{1}{2}E_4(\alpha_A^* - \xi_A^*(t))^2 + E_5(\alpha_S^* - \xi_S^*(t))(\alpha_A^* - \xi_A^*(t)) + E_6(\alpha_S - \xi_S(t))(\alpha_A^* - \xi_A^*(t)) + \text{c.c.}\right]\right\} \tag{73}$$

which generalizes the Kárská and Peřina result for antinormal ordering[56] going over into s-ordering of the field. The time-dependent functions E_i $(i = 1, \ldots, 6)$ and $L^{(1)}$ have been calculated by Peřinová[47] in her analysis of quadratic optical parametric processes. Here, we have the following generalized s-parametrized functions E_i and $L^{(s)}$ occurring in (73):

$$
\begin{aligned}
E_1 &= B_S^{(s)}(t) K_A^{(s)}(t) - B_A^{(s)}(t) K_+(t) \\
&\quad + \left(C_A^*(t) D_{SA}(t) \bar{D}_{SA}(t) + \text{c.c.}\right) \\
E_2 &= B_A^{(s)}(t) K_S^{(s)}(t) - B_S^{(s)}(t) K_+(t) \\
&\quad + \left(C_S(t) D_{SA}^*(t) \bar{D}_{SA}(t) + \text{c.c.}\right) \\
E_3 &= C_S(t) K_A^{(s)}(t) + 2B_A^{(s)}(t) D_{SA}(t) \bar{D}_{SA}^*(t) \\
&\quad + C_A^*(t) D_{SA}^2(t) + C_A(t) \bar{D}_{SA}^{*2}(t) \\
E_4 &= C_A(t) Q(t) K_S^{(s)}(t) + 2B_S^{(s)}(t) D_{SA}(t) \bar{D}_{SA}(t) \\
&\quad + C_S(t) \bar{D}_{SA}^2(t) + C_S^*(t) D_{SA}^2(t) \\
E_5 &= D_{SA}(t)\left[B_S^{(s)}(t) B_A^{(s)}(t) - K_-(t)\right] \\
&\quad + B_S^{(s)}(t) C_A(t) \bar{D}_{SA}^*(t) + B_A^{(s)}(t) C_S(t) \bar{D}_{SA}(t) \\
&\quad + C_S(t) C_A(t) D_{SA}^*(t) \\
E_6 &= -\bar{D}_{SA}(t)\left[B_S^{(s)}(t) B_A^{(s)}(t) + K_-(t)\right] \\
&\quad - B_S^{(s)}(t) C_A(t) D_{SA}^*(t) - B_A^{(s)}(t) Q(t) C_S^*(t) D_{SA}(t) \\
&\quad - C_S^*(t) C_A(t) \bar{D}_{SA}^*(t)
\end{aligned}
\tag{74}
$$

$$
\begin{aligned}
\left(L^{(s)}\right)^2 &= K_S^{(s)}(t) K_A^{(s)}(t) - 2B_S^{(s)}(t) B_A^{(s)}(t) K_+(t) \\
&\quad + \left[C_S(t) C_A(t) D_{SA}^{*2}(t) + C_S(t) C_A^*(t) \bar{D}_{SA}^2(t)\right. \\
&\quad + 2B_S^{(s)}(t) C_A^*(t) D_{SA}(t) \bar{D}_{SA}(t) \\
&\quad \left. + 2B_A^{(s)}(t) C_S^*(t) D_{SA}^*(t) \bar{D}_{SA}(t) + \text{c.c.}\right] + K_-^2(t)
\end{aligned}
\tag{75}
$$

with

$$
\begin{aligned}
K_{S,A}^{(s)}(t) &= \left(B_{S,A}^{(s)}(t)\right)^2 - \left|C_{S,A}(t)\right|^2 \\
K_\pm(t) &= \left|D_{SA}(t)\right|^2 \pm \left|\bar{D}_{SA}(t)\right|^2
\end{aligned}
\tag{76}
$$

The two-mode functions $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ (73) and $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$ (67) reduce to the single-mode functions $\mathscr{W}^{(s)}(\alpha_k, t)$ and $\mathscr{C}^{(s)}(\beta_k, t)$ $(k = S, A)$ simply by setting either $\alpha_S = \beta_S = 0$ or $\alpha_A = \beta_A = 0$, implying that the coefficients $V_S(t)$, $V_A(t)$, $D_{SA}(t)$, and $\bar{D}_{SA}(t)$ vanish and, for instance, $L^{(s)}$ reduces to $\sqrt{K_k^{(s)}(t)}$.

Contrary to the solution (67) for the characteristic function $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$, the solution (73) for the quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ may be absent for some s-ordering of the field operators, depending on the choice of initial field. The condition for the existence of the QPD (73), i.e., the existence of the Fourier transform (21) of $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$ (67), is that the function $K_A^{(s)}(t)$, $L^{(s)}(t)$, $\operatorname{Re} C_A(t) + B_A^{(s)}(t)$, and

$$\bar{L}^{(s)} \equiv \left(K_A^{(s)}(t)\right)^{1/2}\left[\operatorname{Re} C_S(t) + B_S^{(s)}(t)\right] + \left(K_A^{(s)}(t)\right)^{-1/2} \times\left[\operatorname{Re} C_A^*(t)\left(\bar{D}_{SA}(t) - D_{SA}(t)\right)^2 - B_A^{(s)}(t)\left|\bar{D}_{SA}(t) - D_{SA}(t)\right|^2\right] > 0 \tag{77}$$

should be positive. If any of the four functions $K_A^{(s_1)}(t)$, $L^{(s_1)}(t)$, $\bar{L}^{(s_1)}(t)$, and $\operatorname{Re} C_A(t) + B_A^{(s_1)}(t)$ (for a particular parameter s_1) is not positive definite everywhere, the equation of motion (64) for the s_1-parametrized quasidistribution cannot be interpreted as a FPE describing the Brownian motion, i.e., the equation is not a "true" FPE. The quasidistribution $\mathscr{W}^{(s_1)}(\alpha_S, \alpha_A, t)$ does not exist as a positive well-behaved function; still it does exist as a generalized function according to the Klauder-Sudarshan theorem.[198] This property is a signature of quantum effects.[98-100] Let us note that it is possible to use generalized P-representations (positive P-representations) by doubling the phase space, as has been proposed by Drummond and Gardiner.[124] The generalized P-representations have been applied successfully to solve master equations of various nonlinear problems (see, e.g., Ref. 124, 127, 128, and 200). This method, if applied to our model, requires us to handle eight real variables (not counting time), instead of four.

For initially coherent Stokes and anti-Stokes fields, i.e., satisfying $C_S = C_A = \bar{D}_{SA} = 0$, the rather complicated expressions for $\bar{L}^{(s)}$ (77) and $L^{(s)}$ (75) reduce to

$$\bar{L}^{(s)} = B_S^{(s)}(t) B_A^{(s)}(t) - \left|D_{SA}(t)\right|^2 \tag{78}$$

$$L^{(s)} = \left|B_S^{(s)}(t) B_A^{(s)}(t) - \left|D_{SA}(t)\right|^2\right| \tag{79}$$

It is seen that, in the case of initially coherent fields, the sufficient

condition for the existence of $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ (73) is only that the function $\bar{L}^{(s)}$ (78) shall be positive. One obtains further simplifications of the problem under the assumption of negligible frequency mismatch ($\Delta\Omega = 0$). The functions $B^{(s)}_{S,A}(t)$ and $D_{SA}(t)$ (68) now reduce to

$$\begin{aligned} B^{(s)}_S(t) &= \frac{\kappa_S}{\kappa_S - \kappa_A} f_- \left(\frac{\kappa_S}{\kappa_S - \kappa_A} f_+ - 2\frac{\kappa_A}{\kappa_S - \kappa_A} + \langle \hat{n}_V \rangle f_+ \right) \\ &\quad + \frac{1 - s}{2} \geq 0 \\ B^{(s)}_A(t) &= \frac{\kappa_A}{\kappa_S - \kappa_A} f_- \left(\frac{\kappa_S}{\kappa_S - \kappa_A} f_- + \langle \hat{n}_V \rangle f_+ \right) + \frac{1 - s}{2} \geq 0 \\ |D_{SA}(t)| &= \frac{\sqrt{\kappa_S \kappa_A}}{\kappa_S - \kappa_A} f_- \left(\frac{\kappa_S}{\kappa_S - \kappa_A} f_- + \langle \hat{n}_V \rangle f_+ + 1 \right) \end{aligned} \tag{80}$$

with

$$f_\pm = \exp\left(\frac{\kappa_S - \kappa_A}{2} \Delta t \right) \pm 1 \tag{81}$$

In particular, the Wigner function exists, since

$$\bar{L}^{(0)} = \frac{1}{4} + \frac{1}{2} \frac{\exp[(\kappa_S - \kappa_A)\,\Delta t] - 1}{\kappa_S - \kappa_A} [\langle \hat{n}_V \rangle (\kappa_S + \kappa_A) + \kappa_S] > 0 \tag{82}$$

contrary to the P-function, which does not exist for $t > t_0$, since[53]

$$\bar{L}^{(1)} = -\frac{\kappa_S \kappa_A}{(\kappa_S - \kappa_A)^2} \left[\exp\left(\frac{\kappa_S - \kappa_A}{2} \Delta t \right) - 1 \right]^2 < 0 \qquad \text{for } \Delta t > 0 \tag{83}$$

In general, the solution (73) at a given time t exists for parameters s less than

$$s < B^{(1)}_S(t) + B^{(1)}_A(t) + 1 - \sqrt{\left(B^{(1)}_S(t) + B^{(1)}_A(t) \right)^2 - 4\bar{L}^{(1)}} \tag{84}$$

Assuming that the damping constant γ_A is equal to the gain constant γ_S, or equivalently $\kappa_A = \kappa_S = \kappa$, we arrive at

$$\overline{L}^{(s)} = \tfrac{1}{4}\left[(1-s)^2 + 2(1-s)\left(1 + 2\langle \hat{n}_V \rangle\right)\kappa\,\Delta t - s\kappa^2(\Delta t)^2\right] \tag{85}$$

which is greater than zero for parameters s less than

$$s < \frac{1}{2} + \frac{(\kappa\,\Delta t + 1)^2}{2} + \left\{2\langle \hat{n}_V \rangle - \left[\left(1 + 2\langle \hat{n}_V \rangle + \frac{\kappa\,\Delta t}{2}\right)^2 + 1\right]^{1/2}\right\}\kappa\,\Delta t \tag{86}$$

In particular, $\overline{L}^{(1)}$ (83) for the P-function and $\overline{L}^{(0)}$ (82) for the Wigner function respectively reduce to

$$\begin{aligned} \overline{L}^{(1)} &= -\left(\frac{\kappa\,\Delta t}{2}\right)^2 < 0 \qquad \text{for } \Delta t > 0 \\ \overline{L}^{(0)} &= \tfrac{1}{4} + \left(\langle \hat{n}_V \rangle + \tfrac{1}{2}\right)\kappa\,\Delta t > 0 \end{aligned} \tag{87}$$

The condition for s fulfilling $\overline{L}^{(s)} > 0$ cannot be expressed explicitly in a simple form in cases with frequency mismatch $\Delta\Omega \neq 0$. As another example, let us assume that the Stokes and anti-Stokes fields are initially chaotic, which mathematically differs from our former example of initially coherent state by the presence of nonzero initial coefficients $B_k^{(-1)} = \langle \hat{n}_{\mathrm{ch}\,k} \rangle$ $(k = S, A)$. By virtue of the relations (68), the functions $B_k^{(s)}(t)$, $D_{SA}(t)$ for chaotic field are the same as for a coherent field with extra terms. Here, the function $\overline{L}^{(s)}$ (75) is found to be

$$\begin{aligned} \overline{L}^{(s)} = &\left(B_S^{(s)}(t) + \langle \hat{n}_{\mathrm{ch}\,S} \rangle |U_S(t)|^2 + \langle \hat{n}_{\mathrm{ch}\,A} \rangle |V_S(t)|^2\right) \\ &\times \left(B_A^{(s)}(t) + \langle \hat{n}_{\mathrm{ch}\,A} \rangle |U_A(t)|^2 + \langle \hat{n}_{\mathrm{ch}\,S} \rangle |V_A(t)|^2\right) \\ &+ \left| |D_{SA}| + \langle \hat{n}_{\mathrm{ch}\,S} \rangle U_S(t) V_A(t) + \langle \hat{n}_{\mathrm{ch}\,A} \rangle U_A(t) V_S(t)\right|^2 \end{aligned} \tag{88}$$

which has a form similar to (79) with the same function $B_k^{(s)}(t)$ and $D_{SA}(t)$

given by (80). In the case of equal damping and gain constants we obtain

$$\begin{aligned}\bar{L}^{(s)} = &-\left(\frac{\gamma\Delta t}{2}\right)^2 s\left(\langle\hat{n}_{\mathrm{ch}\,A}\rangle + \langle\hat{n}_{\mathrm{ch}\,S}\rangle - 1\right)\\ &+\gamma\Delta t\left[\langle\hat{n}_{\mathrm{ch}\,A}\rangle\left(\langle\hat{n}_V\rangle + \frac{1+s}{2}\right)\right.\\ &\left.+\langle\hat{n}_{\mathrm{ch}\,S}\rangle\left(\langle\hat{n}_V\rangle + \frac{1-s}{2}\right) + \left(\langle\hat{n}_V\rangle + \frac{1}{2}\right)(1-s)\right]\\ &+\langle\hat{n}_{\mathrm{ch}\,A}\rangle\left(\langle\hat{n}_{\mathrm{ch}\,S}\rangle + \frac{1-s}{2}\right) + \langle\hat{n}_{\mathrm{ch}\,S}\rangle\frac{1-s}{2} + \left(\frac{1-s}{2}\right)^2\end{aligned} \tag{89}$$

It is seen that the Wigner function always exists, since

$$\begin{aligned}\bar{L}^{(0)} = &\gamma\Delta t\left(\langle\hat{n}_{\mathrm{ch}\,S}\rangle + \langle\hat{n}_{\mathrm{ch}\,A}\rangle + 1\right)\left(\langle\hat{n}_V\rangle + \tfrac{1}{2}\right)\\ &+\langle\hat{n}_{\mathrm{ch}\,A}\rangle\left(\langle\hat{n}_{\mathrm{ch}\,S}\rangle + \tfrac{1}{2}\right) + \tfrac{1}{2}\langle\hat{n}_{\mathrm{ch}\,S}\rangle + \tfrac{1}{4} > 0\end{aligned} \tag{90}$$

whereas the P-function exists only for times shorter than

$$\begin{aligned}\Delta t < \frac{2}{\gamma}&\left(\langle\hat{n}_{\mathrm{ch}\,A}\rangle + \langle\hat{n}_{\mathrm{ch}\,S}\rangle + 1\right)^{-1}\left\{\langle\hat{n}_{\mathrm{ch}\,A}\rangle^2\left[\langle\hat{n}_{\mathrm{ch}\,S}\rangle + \left(\langle\hat{n}_V\rangle + 1\right)^2\right]\right.\\ &+\langle\hat{n}_{\mathrm{ch}\,A}\rangle\langle\hat{n}_{\mathrm{ch}\,S}\rangle\left[\langle\hat{n}_{\mathrm{ch}\,S}\rangle + \langle\hat{n}_V\rangle^2 + \left(\langle\hat{n}_V\rangle + 1\right)^2\right]\\ &\left.+\langle\hat{n}_{\mathrm{ch}\,S}\rangle^2\langle\hat{n}_V\rangle^2\right\}^{1/2}\\ &+\langle\hat{n}_{\mathrm{ch}\,A}\rangle\left(\langle\hat{n}_V\rangle + 1\right) + \langle\hat{n}_{\mathrm{ch}\,S}\rangle\langle\hat{n}_V\rangle\end{aligned} \tag{91}$$

The relation (89) is quadratic in s and readily gives an analytic expression for the largest parameter s ($s \leq 1$) for which the quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ exists at a given time of the evolution $\Delta t = t - t_0$.

In Fig. 1 we present the function $\bar{L}^{(s)}(t)$ for different values of the frequency mismatch $\Delta\Omega$, of the mean number of photons $\langle\hat{n}_V\rangle$, and of the damping (κ_A) and gain (κ_S) constants. We assume that the Stokes and anti-Stokes modes are initially coherent. Thus, for all discussed cases (Figs. 1a–d), the condition of a positive definite function $\bar{L}^{(s)}(t)$ is sufficient for the existence of the corresponding s-parametrized QPD. For clarity, the dashed lines in Fig. 1 are depicted for $\bar{L}^{(s)}(t) = 0$.

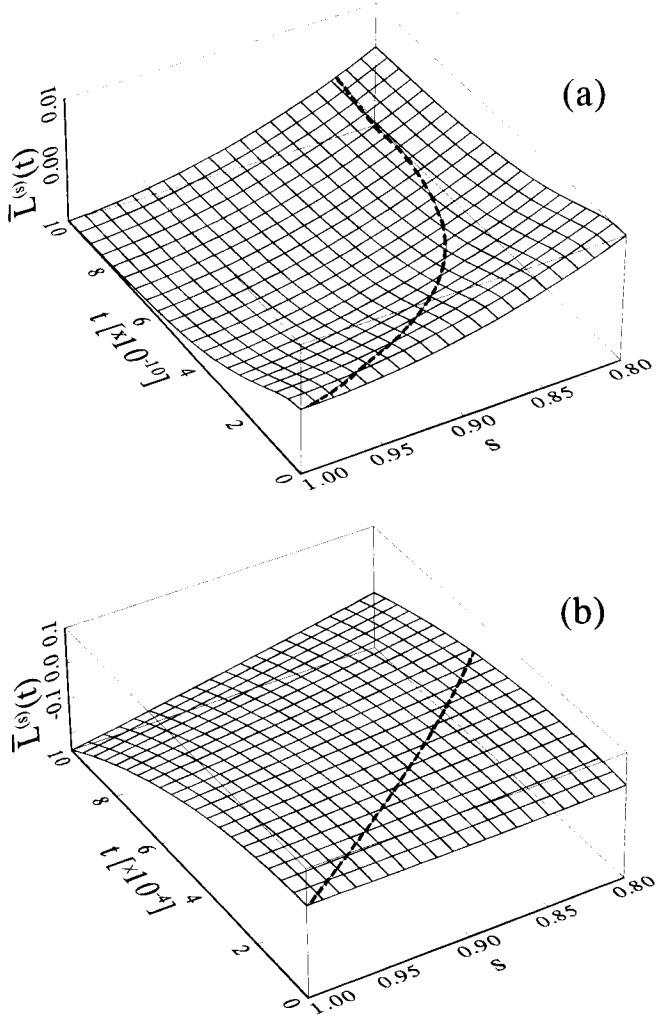

Figure 1a. The time and parameter s dependence of the function $\bar{L}^{(s)}(t)$, related to the existence of the QPD $W^{(s)}(\alpha_S, \alpha_A, t)$, for (a) $\kappa_S = 10^8$, $\kappa_A = 10^{10}$, $|\Delta\Omega| = 1 \div 10^6$ (the surfaces coincide in this range of $|\Delta\Omega|$), $\langle \hat{n}_V \rangle = 0$; (b) $\kappa_S = \kappa_A = 10^3$, $|\Delta\Omega| = 1$, $\langle \hat{n}_V \rangle = 0 \div 100$; (c) $\kappa_S = \kappa_A = 10^8$, $|\Delta\Omega| = 10^6$, $\langle \hat{n}_V \rangle = 10$; and (d) $\kappa_S = \kappa_A = 10^8$, $|\Delta\Omega| = 10^6$, $\langle \hat{n}_V \rangle = 0$. The Stokes and anti-Stokes fields are initially coherent. The dashed lines on the surfaces are depicted for $\bar{L}^{(s)}(t) = 0$.

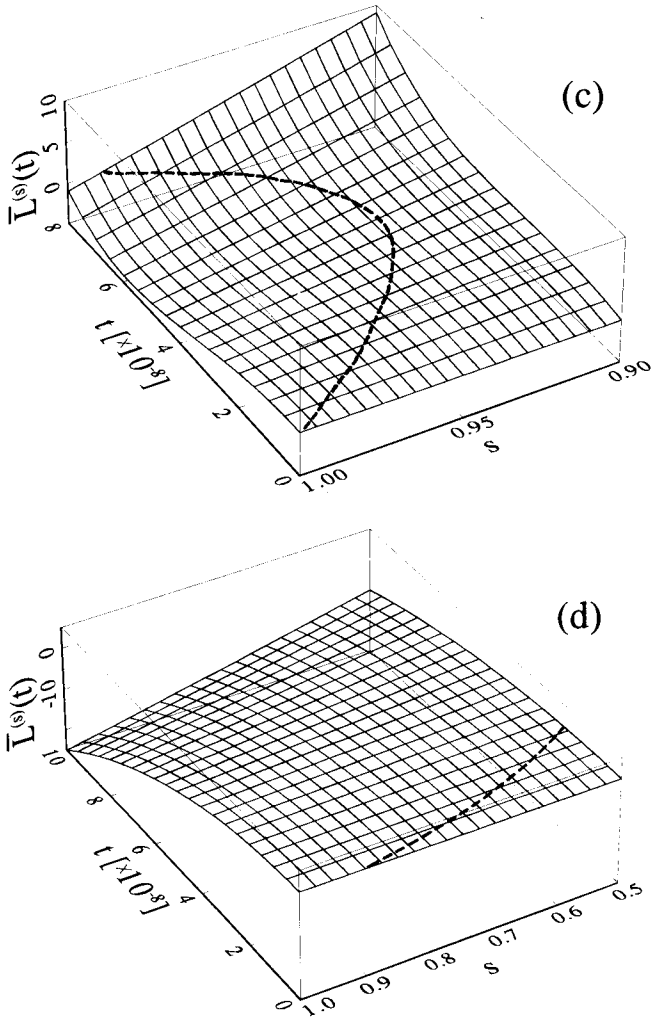

Figure 1b. *(Continued)*

We shall briefly analyze a more general situation, which comprises the above cases and others. Let us assume after Refs. 56 and 66 (for a general analysis see Refs. 23 and 219) that the Stokes ($k = S$) as well anti-Stokes (A) modes are initially in squeezed states characterized by complex amplitudes ξ_k, parameters r_k, and phases ϕ_k, superposed with a chaotic field, characterized by the mean number of chaotic photons $\langle \hat{n}_{\mathrm{ch}\,k} \rangle$. The

initial s-parametrized quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t_0)$ is then given by

$$\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t_0) = \prod_{k=S,A} \left(K_k^{(s)}\right)^{-1/2} \times \exp\left\{ -\frac{1}{K_k^{(s)}} \left[B_k^{(s)} |\alpha_k - \xi_k|^2 - \mathrm{Re}\left(C_k^* (\alpha_k - \xi_k)^2\right) \right] \right\} \tag{92}$$

with

$$B_k^{(s)} = B_k^{(s)}(t_0) = (\cosh r)^2 + \langle n_{\mathrm{ch}\,k} \rangle - \frac{s+1}{2}$$

$$C_k = C_k(t_0) = \tfrac{1}{2} \exp(\mathrm{i}\phi_k) \sinh(2r_k) \tag{93}$$

which trivially reduces to the quasidistributions of a pure squeezed state ($\langle \hat{n}_{\mathrm{ch}\,k} \rangle = 0$), a coherent state ($\langle \hat{n}_{\mathrm{ch}\,k} \rangle = r_k = 0$), or a chaotic state ($r_k = \xi_k = 0$). In Section VI.B we analyze another special case of (93) with $r_k = 0$ describing a general superposition of coherent and chaotic fields.

The Raman effect model under parametric approximation is fully specified either by the s-parametrized characteristic function $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$ (67) or the s-parametrized quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ (73). In particular, by virtue of the relations presented in Section IV, one can obtain complete information about the photon-counting statistics and squeezing properties of the scattered fields from (67) or (73).

One can calculate the photon-counting probability distribution $p(n)$ from the quasidistribution $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ or integrated quasidistribution $\mathscr{W}^{(s)}(W, t)$ (30) by means of (31), or equivalently from the generating function $\langle \exp(-\lambda W(t)) \rangle_{(s)}$ (32) by virtue of (33). We apply the latter method, which gives us, after insertion of $\mathscr{W}^{(s)}(\alpha_S, \alpha_A, t)$ (73) or $\mathscr{C}^{(s)}(\beta_S, \beta_A, t)$ (67) into (32), the following time-dependent s-parametrized generating function:

$$\langle \exp(-\lambda W) \rangle_{(s)} = \lambda^{-2} \left(\mathscr{L}_1^{(s)}\right)^{-1/2} \exp\left(\frac{\mathscr{L}_2^{(s)}}{\mathscr{L}_1^{(s)}} \right) \tag{94}$$

where the $\mathscr{L}_1^{(s)}$ ($\mathscr{L}_2^{(s)}$) are polynomials of the fourth (third) order in λ^{-1}:

$$\mathscr{L}_1^{(s)} = \sum_{j=0}^{4} \left(\lambda^{-1} + \frac{1-s}{2}\right)^j b_j$$

$$\mathscr{L}_2^{(s)} = \sum_{j=0}^{3} \left(\lambda^{-1} + \frac{1-s}{2}\right)^j a_j \tag{95}$$

Adapting the results of Peřinová and Peřina[48] for the coefficients a_j, b_j ($j = 0, 1, \ldots$) occurring in (95), one obtains

$$\begin{aligned}
a_0 &= \left[-B_S^{(1)}K_A^{(1)} + B_A^{(1)}K_+ + \left(C_A D_{SA}\overline{D}_{AS} + \text{c.c.}\right)\right]|\xi_S|^2 \\
&\quad + \Big\{\left[B_A^{(1)}D_{SA}\overline{D}_{SA} + \tfrac{1}{2}\left(C_S^*K_A^{(1)} + C_A D_{SA}^2 + C_A^*\overline{D}_{SA}^2\right)\right]\xi_S^2 \\
&\qquad + \tfrac{1}{2}\Big[B_S^{(1)}B_A^{(1)}D_{SA} + 2B_S^{(1)}C_A^*\overline{D}_{SA} + C_S^*C_A^*D_{SA}^* - D_{SA}K_-\Big]\xi_S\xi_A \\
&\qquad\quad - \tfrac{1}{2}\Big[B_S^{(1)}B_A^{(1)}\overline{D}_{SA} + 2B_S^{(1)}C_A D_{SA} + C_S^*C_A\overline{D}_{AS} \\
&\qquad\qquad + \overline{D}_{SA}K_-\Big]\xi_S\xi_A^* + \text{c.c.}\Big\} + [S \leftrightarrow A] \\
a_1 &= \left[-2B_S^{(1)}B_A^{(1)} - K_A^{(1)} + K_+\right)|\xi_S|^2 \\
&\quad + \Big[\left(B_A^{(1)}C_S^* + D_{SA}\overline{D}_{SA}\right)\xi_S^2 + \left(B_S^{(1)}D_{SA} + C_S^*\overline{D}_{AS}\right)\xi_S\xi_A \\
&\qquad - \left(B_S^{(1)}\overline{D}_{SA} + C_S^*D_{SA}^*\right)\xi_S\xi_A^* + \text{c.c.}\Big] \\
&\quad + [S \leftrightarrow A] \\
a_2 &= -\left(B_S^{(1)} + 2B_A^{(1)}\right)|\xi_S|^2 \\
&\quad + \tfrac{1}{2}\left(C_S^*\xi_S^2 + D_{SA}\xi_S\xi_A - \overline{D}_{SA}\xi_S\xi_A^* + \text{c.c.}\right) + [S \leftrightarrow A] \\
a_3 &= -|\xi_S|^2 - |\xi_A|^2 \\
b_0 &= \tfrac{1}{2}K_S^{(1)}K_A^{(1)} - B_S^{(1)}B_A^{(1)}K_+ - 2B_S^{(1)}\left(C_A^*D_{SA}^*\overline{D}_{AS} + \text{c.c.}\right) + \tfrac{1}{2}K_-^2 \\
&\quad - \tfrac{1}{2}\Big[C_A\left(C_S D_{SA}^2 + C_S^*\overline{D}_{AS}^2\right) + \text{c.c.}\Big] + [S \leftrightarrow A] \\
b_1 &= 2B_S^{(1)}\left(K_A^{(1)} - K_+\right) - 2\left(C_S D_{SA}\overline{D}_{SA} + \text{c.c.}\right) + [S \leftrightarrow A] \\
b_2 &= 2B_S^{(1)}B_A^{(1)} + K_S^{(1)} - K_+ + [S \leftrightarrow A] \\
b_3 &= 2\left(B_S^{(1)} + B_A^{(1)}\right) \\
b_4 &= 1
\end{aligned} \tag{96, 97}$$

where $[S \leftrightarrow A]$ stands for the preceding terms albeit with interchanged subscripts S and A. For brevity, we have omitted the time dependence of Eqs. (96) and (97). Our formulas (94) and (95) are generalizations of the results given in Refs. 23, 48, and 56 for $s = -1$ to any s. It is seen that the simplest form of (95) is for normal ordering of the field operators; hence, here, we use only this ordering. Peřinová and Peřina[48] have shown that if the polynomial $\mathscr{L}_1^{(1)}$ has four single roots $\lambda_k = -(1/\lambda)_k$, the generating function $\langle \exp(-\lambda W(t))\rangle_{(1)}$ has the form of the fourfold generating function for Laguerre polynomials

$$\langle \exp(-\lambda W(t))\rangle_{(1)} = \prod_{k=1}^{4} (1 + \lambda\lambda_k)^{-1/2} \exp\left(-\frac{\lambda A_k}{1 + \lambda\lambda_k}\right) \tag{98}$$

The field is described by a superposition of signal components

$$A_k = \prod_{\substack{l=1 \\ l\neq k}}^{4} \left(\lambda_k^{-1} - \lambda_l^{-1}\right)^{-1} \sum_{l=0}^{3} a_l \left(-\lambda_k^{-1}\right)^l \tag{99}$$

and the noise components λ_k. With Eq. (98) available, one obtains[48, 56] the following photocount distribution

$$p(n,t) = \sum_{\substack{k_1,k_2,k_3,k_4 \\ k_1+k_2+k_3+k_4=n}} \prod_{l=1}^{4} \exp\left(-\frac{A_l}{1+\lambda_l}\right) \times \frac{\lambda_l^{k_l}}{(1+\lambda_l)^{k_l+1/2}\Gamma\left(k_l+\frac{1}{2}\right)} L_{k_l}^{-1/2}\left(\frac{-A_l}{\lambda_l(1+\lambda_l)}\right) \tag{100}$$

and its factorial moments

$$\langle W^k(t)\rangle_{(1)} = k! \sum_{k_1+k_2+k_3+k_4=k} \prod_{l=1}^{4} \frac{\lambda_l^{k_l}}{\Gamma\left(k_l+\frac{1}{2}\right)} L_{k_l}^{-1/2}\left(-\frac{A_l}{\lambda_l}\right) \tag{101}$$

by applying well-known properties of the generating function of the generalized Laguerre polynomials $L_k^{\alpha}(x)$ to the definition relations (33) for $p(n,t)$ with $s = 1$ and to the relations (34) for $\langle W^k(t)\rangle_{(1)}$. Much simpler expressions are found in the special case when the radiation fields are initially superpositions of coherent and chaotic fields. From relations (93) with $r_k = 0$ and (68) it is seen that $C_S(t) = C_A(t) = \bar{D}_{SA}(t) = 0$. The fourfold generating function (98) reduces to a twofold generating function

in the form of (98), where the upper limit of the product should be replaced by 2 and the square root in $(1 + \lambda\lambda_k)^{-1/2}$ should be omitted. Then the photocount distribution $p(n, t)$ and its factorial moments $\langle W^k(t)\rangle_{(1)}$ become

$$p(n,t) = (n!)^{-1}\exp\left(-\frac{A_1}{1+\lambda_1} - \frac{A_2}{1+\lambda_2}\right)\sum_{l=0}^{n}\binom{n}{l}\lambda_1^l\lambda_2^{n-l}(1+\lambda_1)^{-(l+1)}$$
$$\times(1+\lambda_2)^{-n+(l+1)}L_l\left(-\frac{A_1}{\lambda_1(1+\lambda_1)}\right)L_{n-1}\left(-\frac{A_2}{\lambda_2(1+\lambda_2)}\right) \tag{102}$$

$$\langle W^n(t)\rangle_{(1)} = \sum_{l=0}^{n}\binom{n}{l}\lambda_1^l\lambda_2^{n-l}L_l\left(-\frac{A_1}{\lambda_1}\right)L_{n-l}\left(-\frac{A_2}{\lambda_2}\right) \tag{103}$$

where $L_n(x) = L_n^0(x)$ and the roots λ_k and coefficients A_k are[48]

$$\lambda_{1,2} = \tfrac{1}{2}\left\{B_S^{(1)}(t) + B_A^{(1)}(t) \mp \left[\left(B_S^{(1)}(t) - B_A^{(1)}(t)\right)^2 + 4\left|D_{SA}(t)\right|^2\right]^{1/2}\right\}$$
$$A_{1,2} = \pm\left[\left(B_S^{(1)}(t) - B_A^{(1)}(t)\right)^2 + 4\left|D_{SA}(t)\right|^2\right]^{-1/2} \tag{104}$$
$$\times\left[\tfrac{1}{2}\left(B_S^{(1)}(t) - B_A^{(1)}(t)\right)\left(|\xi_A|^2 - |\xi_S|^2\right) - \left(D_{SA}\xi_S\xi_A^* + \text{c.c.}\right)\right]$$
$$+\tfrac{1}{2}\left(|\xi_S|^2 + |\xi_A|^2\right)$$

The photon-counting statistics of scattering either into the Stokes or anti-Stokes mode can be calculated from formulas (98)–(101). In the single-mode case the moments $D_{SA}(t)$ and $\bar{D}_{SA}(t)$ vanish, considerably simplifying the polynomial $\mathscr{L}_1^{(1)}$ (95), with coefficients b_j, to the form

$$\mathscr{L}_1^{(1)} = \prod_{k=S,A}\left(\lambda^{-2} + 2\lambda^{-1}B_k^{(1)}(t) + K_k^{(1)}(t)\right) \tag{105}$$

with the roots $\lambda_{1,2\,S,A} = -(\lambda^{-1})_k$

$$\lambda_{1,2k} = B_k^{(1)}(t) \mp |C_k(t)| \qquad (k = S, A) \tag{106}$$

The notation $\lambda_{1S,A}$ and $\lambda_{2S,A}$, instead of $\lambda_{1,2,3,4}$, emphasizes the dependence on the single-mode variables in accordance with the assumption of

alternative scattering into the Stokes or anti-Stokes mode. Analogously, it is seen that

$$A_{1,2k} = \tfrac{1}{2}|\xi_k(t)|^2 \mp \tfrac{1}{4}|C_k(t)|^{-1}\left(C_k^*(t)\xi_k^2(t) + \text{c.c}\right) \tag{107}$$

On insertion of (106) into twofold functions (98)–(101) one immediately obtains

$$\begin{aligned}\left\langle \exp(-\lambda W_k)\right\rangle_{(1)} &= \left[(1+\lambda\lambda_{1k})(1+\lambda\lambda_{2k})\right]^{-1/2} \\ &\quad \times \exp\left[-\lambda A_{1k}(1+\lambda\lambda_{1k})^{-1} - \lambda A_{2k}(1+\lambda\lambda_{2k})^{-1}\right]\end{aligned} \tag{108}$$

$$\begin{aligned}p_k(n) &= \left[(1+\lambda_{1k})(1+\lambda_{2k})\right]^{-1/2}\left(1+\lambda_{2k}^{-1}\right)^{-n} \\ &\quad \times \exp\left(-\frac{A_{1k}}{1+\lambda_{1k}} - \frac{A_{2k}}{1+\lambda_{2k}}\right) \\ &\quad \times \sum_{l=0}^{n} \frac{1}{\Gamma(l+\frac{1}{2})\Gamma(n-l+\frac{1}{2})}\left(\frac{1+\lambda_{2k}^{-1}}{1+\lambda_{1k}^{-1}}\right)^{l} \\ &\quad \times L_l^{-1/2}\left(-\frac{A_{1k}}{\lambda_{1k}(1+\lambda_{1k})}\right) L_{n-l}^{-1/2}\left(-\frac{A_{2k}}{\lambda_{2k}(1+\lambda_{2k})}\right)\end{aligned} \tag{109}$$

$$\begin{aligned}\left\langle W_k^n(t)\right\rangle_{(1)} &= n!\lambda_{2k}^n \sum_{l=0}^{n} \frac{1}{\Gamma(l+\frac{1}{2})\Gamma(n-l+\frac{1}{2})}\left(\frac{\lambda_{1k}}{\lambda_{2k}}\right)^{l} \\ &\quad \times L_l^{-1/2}\left(-\frac{A_{1k}}{\lambda_{1k}}\right) L_{n-l}^{-1/2}\left(-\frac{A_{2k}}{\lambda_{2k}}\right)\end{aligned} \tag{110}$$

To obtain the results of Refs. 23, 56, and 189, one should replace λ_{1k} by $E_k - 1$ and λ_{2k} by $F_k - 1$. In particular, assuming that a scattered (Stokes or anti-Stokes) mode is initially in a coherent state (thus $C_k = 0$) the mean photon numbers $\langle \hat{n}_k \rangle$ ($k = S, A$) are

$$\left\langle \hat{n}_k(t)\right\rangle = \left\langle W_k(t)\right\rangle_{(1)} = |\xi_k(t)|^2 + B_k^{(1)}(t) \tag{111}$$

or explicitly

$$\left\langle \hat{n}_S(t)\right\rangle = |\xi_S|^2 \exp(\kappa_S \Delta t) + \left(\langle \hat{n}_V \rangle + 1\right)\left[\exp(\kappa_S \Delta t) - 1\right] \tag{112}$$

$$\left\langle \hat{n}_A(t)\right\rangle = |\xi_A|^2 \exp(-\kappa_A \Delta t) + \langle \hat{n}_V \rangle\left[1 - \exp(-\kappa_A \Delta t)\right] \tag{113}$$

whereas the mean-square photon-numbers $\langle \hat{n}_k^2 \rangle$ are

$$\begin{aligned}\langle \hat{n}_k^2 \rangle &= \langle W_k^2 \rangle_{(1)} + \langle W_k \rangle_{(1)} \\ &= |\xi_k(t)|^4 + |\xi_k(t)|^2 (4B_k(t) + 1) + 2B_k^2(t) + B_k(t)\end{aligned} \tag{114}$$

Then, the normalized second-order factorial moments (35) are equal to

$$\begin{aligned}\gamma_k^{(2)}(t) = B_k^{(1)}(t)\left[|\xi_k(t)|^2 + B_k^{(1)}(t)\right]^{-1} \\ \times \left\{ |\xi_k(t)|^2 \left[|\xi_k(t)|^2 + B_k^{(1)}(t)\right]^{-1} + 1 \right\}\end{aligned} \tag{115}$$

Let us proceed to analyze squeezing along the lines presented in Section IV. We focus our attention on single- and two-mode squeezed light according to the definition of "usual" squeezing and principal squeezing of Lukš et al.[132, 211, 213] Using the definitions (66) of the functions $B_k^{(s)}(t)$, $D_{kl}(t)$, $\bar{D}_{kl}(t)$, and $C_k(t)$ we readily obtain expressions for the moments of the quadratures $\hat{X}_k$ and $\hat{X}_{kl}$

$$\left\langle \left(\Delta \hat{X}_{k1,k2}\right)^2 \right\rangle = \pm 2 \operatorname{Re} C_k(t) + 2B_k^{(s)}(t) + s \tag{116}$$

$$\left\langle \left(\Delta \hat{X}_{k\pm}\right)^2 \right\rangle = \pm 2 |C_k(t)| + 2B_k^{(s)}(t) + s \tag{117}$$

$$\begin{aligned}\left\langle \left\{ \Delta \hat{X}_{k1}, \Delta \hat{X}_{k2} \right\} \right\rangle &= 4 \operatorname{Im} C_k(t) \\ \langle \Delta \hat{X}_{k1} \Delta \hat{X}_{l1} \rangle &= 2 \operatorname{Re}\left[D_{kl}(t) - \bar{D}_{kl}(t) \right] \\ \langle \Delta \hat{X}_{k2} \Delta \hat{X}_{l2} \rangle &= -2 \operatorname{Re}\left[D_{kl}(t) + \bar{D}_{kl}(t) \right] \\ \langle \Delta \hat{X}_{k1} \Delta \hat{X}_{l2} \rangle &= 2 \operatorname{Im}\left[D_{kl}(t) - \bar{D}_{kl}(t) \right] \\ \langle \Delta \hat{X}_{k2} \Delta \hat{X}_{l1} \rangle &= 2 \operatorname{Im}\left[D_{kl}(t) + \bar{D}_{kl}(t) \right]\end{aligned} \tag{118}$$

where, as usual, $k, l = S, A$ and $k \neq l$. Thus, the two-mode quadrature variances now have the form

$$\begin{aligned}\left\langle \left(\Delta \hat{X}_{SA1}\right)^2 \right\rangle \\ \left\langle \left(\Delta \hat{X}_{SA2}\right)^2 \right\rangle\end{aligned} = \pm 2 \operatorname{Re}\left[C_S(t) + C_A(t) + 2D_{SA}(t) \right] + 2\left[B_S^{(s)}(t) + B_A^{(s)}(t) - 2 \operatorname{Re} \bar{D}_{SA}(t) + s \right] \tag{119}$$

and the extremal variances are

$$\left\langle \left(\Delta \hat{X}_{SA\pm}\right)^2 \right\rangle = \pm 2\left|C_S(t) + C_A(t) + 2D_{SA}(t)\right| + 2\left[B_S^{(s)}(t) + B_A^{(s)}(t) - 2\,\mathrm{Re}\,\overline{D}_{SA}(t) + s\right] \tag{120}$$

The single-mode squeezing, defined in standard manner, and the single-mode principal squeezing require, respectively, that

$$\left.\begin{matrix} |\mathrm{Re}\, C_k(t)| \\ |C_k(t)| \end{matrix}\right\rangle > B_k^{(s)}(t) + \frac{s}{2} \qquad (k = S, A) \tag{121}$$

whereas the conditions for the two-mode squeezing are, respectively,

$$\left.\begin{matrix} |\mathrm{Re}[C_S(t) + C_A(t) + 2D_{SA}(t)]| \\ |C_S(t) + C_A(t) + 2D_{SA}(t)| \end{matrix}\right\rangle > B_S^{(s)}(t) + B_A^{(s)}(t) - 2\,\mathrm{Re}\,\overline{D}_{SA}(t) + s \tag{122}$$

Examples of the time evolution of $\langle \hat{n}_S(\tau)\rangle$, $\langle \hat{n}_S^2(\tau)\rangle$, $\langle \hat{a}_S(\tau)\rangle$, and $\langle \hat{a}_S^2(\tau)\rangle$ are given by curves C in Figs. 2, 3, 7, and 8, respectively. We assume that the Stokes fields are initially in a coherent state (stimulated Raman scattering) or in a vacuum state (spontaneous Raman scattering). The rescaled time τ is defined by $t \to \tau = t\gamma_S$. Anti-Stokes scattering is neglected. The phonon bath is at very low temperature, so we put $\langle \hat{n}_V\rangle = 0$. In Fig. 9 we present the time evolution of the extremal variances $\langle (\Delta X_{S\pm}(\tau))^2\rangle$ for fields initially coherent with amplitudes equal to $\alpha_L = \sqrt{2}$, $\alpha_S = \sqrt{0.2}$ and assuming that the heat bath is "quiet" (i.e., $\langle \hat{n}_V\rangle = 0$). In the model under discussion, the variance for the Stokes mode, $\langle (\Delta X_S(\theta, \tau))^2\rangle$ (curve C in Fig. 9), is independent of θ, i.e., $\langle (\Delta X_{S+}(\tau))^2\rangle = \langle (\Delta X_{S-}(\tau))^2\rangle$. Hence, squeezing is not observed if the initial Stokes mode is in a coherent state. Even if the Stokes field is initially squeezed and $\gamma_S > \gamma_A$ (not necessarily $\gamma_A = 0$), squeezing will rapidly vanish due to strong amplification of this mode, which leads to a strong increase in quantum noise.[56] The results of this section (curves C) are compared with the exact solutions (without parametric approximation) derived in Section VI.A.2. (curves A) and the short-time solutions of SectionVI.A.1.

VI. MASTER EQUATION IN FOCK REPRESENTATION

The parametric approximation, applied in the previous section, introduces linearization into our Raman scattering model described by the Hamiltonians (1)–(3). Here, we shall search for a solution to the nonlinear problem, thus including pump depletion. The generalized Fokker-Planck equation (61) and the corresponding equation of motion (63) for the characteristic function reveal the difficulties to be overcome in the complete analysis of Raman scattering into simultaneously both the Stokes and anti-Stokes fields from phonons treated as a "noisy" ($\langle \hat{n}_V \rangle \neq 0$) reservoir. Let us assume that the temperature of the medium is low. Under this assumption it is quite reasonable to neglect the anti-Stokes scattering ($\gamma_A = \gamma_{SA} = \gamma_{AS} = 0$) and, with regard to Eq. (14), to assume that the reservoir is "quiet" ($\langle \hat{n}_V \rangle = 0$). Under these approximations the master equation (18) reduces to the simple form[73]:

$$\frac{\partial \hat{\rho}}{\partial \tau} = \frac{1}{2}\left(\left[\hat{a}_L \hat{a}_S^+, \hat{\rho}\hat{a}_L^+ \hat{a}_S\right] + \left[\hat{a}_L \hat{a}_S^+ \hat{\rho}, \hat{a}_L^+ \hat{a}_S\right]\right) \tag{123}$$

where we have introduced a rescaled time $t \to \tau = \gamma_S t$. Let us denote the matrix elements of the reduced density operator $\hat{\rho}$ in Fock representation by

$$\langle n_L, n_S | \hat{\rho}(\tau) | n'_L, n'_S \rangle \equiv \langle n, m | \hat{\rho}(\tau) | n + \nu, m + \mu \rangle \equiv \rho_{n,m}(\nu, \mu, \tau) \tag{124}$$

where for simplicity we identify $n_L = n$, and $n_S = m$; μ is the degree of off-diagonality for the elements of the matrix $\hat{\rho}$ for the Stokes mode, whereas ν is the degree of off-diagonality for the pump laser mode elements. The master equation for the matrix elements (124) readily follows from Eq. (123) and can be written as

$$\begin{aligned}\frac{\partial}{\partial \tau}\rho_{nm}(\nu\mu\tau) = &-\frac{1}{2}[n(m+1) + (n+\nu)(m+\mu+1)]\rho_{nm}(\nu\mu\tau) \\ &+ [(n+1)(n+\nu+1)m(m+\mu)]^{1/2}\rho_{n+1,m-1}(\nu\mu\tau)\end{aligned} \tag{125}$$

The equation (125) for the diagonal matrix elements $\rho_{nm}(00\tau)$ reduces to the rate equations of Loudon,[30] and McNeil and Walls.[73] Simaan[75] (cf. Ref. 30) analyzed Raman scattering from a gas of two-level atoms. On

the assumption that almost all the atoms are in their ground state, the Simaan rate equation of Ref. 75 takes the form of Eq. (125) for $\nu = \mu = 0$.

A. Raman Scattering Including Pump Depletion

1. *Short-time Solutions*

Before proceeding to derive an exact solution of (125) we shall present the short-time solutions calculated with the help of the relation $\langle \hat{A}(\tau)\rangle = \mathrm{Tr}\{\hat{A}[\hat{\rho}(\tau_0) + \hat{\rho}'(\tau_0)\,\Delta\tau + \hat{\rho}''(\tau_0)(\Delta\tau)^2/2]\}$, where $\hat{\rho}''(\tau_0)$ is found by differentiating Eq. (125) with respect to τ. The solutions for the mean $\langle \hat{n}\rangle$ and mean-square number of photons $\langle \hat{n}^2\rangle$ in the laser mode up to $\Delta\tau$ squared are

$$\langle \hat{n}(\tau)\rangle = \langle \hat{n}\rangle - \langle \hat{n}\rangle(\langle \hat{m}\rangle + 1)\,\Delta\tau - \left[\langle \hat{n}^2\rangle(\langle \hat{m}\rangle + 1) - \langle \hat{n}\rangle(\langle \hat{m}^2\rangle + 3\langle \hat{m}\rangle + 2)\right]\frac{(\Delta\tau)^2}{2} \tag{126}$$

$$\langle \hat{n}^3(\tau)\rangle = \langle \hat{n}^2\rangle - (2\langle \hat{n}^2\rangle - \langle \hat{n}\rangle)(\langle \hat{m}\rangle + 1)\,\Delta\tau - \left[2\langle \hat{n}^2\rangle(\langle \hat{m}\rangle + 1) - \langle \hat{n}^2\rangle(4\langle \hat{m}^2\rangle + 13\langle \hat{m}\rangle + 9) + 3\langle \hat{n}\rangle(\langle \hat{m}^2\rangle + 3\langle \hat{m}\rangle + 2)\right]\frac{(\Delta\tau)^2}{2} \tag{127}$$

where for brevity we set $\langle \hat{n}^p(\tau_0)\rangle = \langle \hat{n}^p\rangle$ and $\langle \hat{m}^p(\tau_0)\rangle = \langle \hat{m}^p\rangle$ ($k = 1, 2, 3$) as well as $\Delta\tau = \tau - \tau_0$). Then the normalized second-order factorial moment, $\gamma_L^{(2)}(\tau)$, defined by (35), is equal to

$$\gamma_L^{(2)}(\tau) = \gamma_L^{(2)} + \left[\left(\langle \hat{n}^2\rangle^2 - \langle \hat{n}^3\rangle\langle \hat{n}\rangle\right)(1 + \langle \hat{m}\rangle) + \langle \hat{n}\rangle\left(\langle \hat{n}^2\rangle - \langle \hat{n}\rangle\right)\left(1 + \langle \hat{m}\rangle - \langle \hat{m}\rangle^2 + \langle \hat{m}^2\rangle\right)\right]\langle \hat{n}\rangle^{-3}(\Delta\tau)^2 \tag{128}$$

which reduces to the Simaan result[75]:

$$\gamma_L^{(2)}(\tau) = \eta_L^{(2)} + \left[\langle \hat{n}^2\rangle\left(\frac{\langle \hat{n}^2\rangle}{\langle \hat{n}\rangle} + 1\right) - \langle \hat{n}^3\rangle - \langle \hat{n}\rangle\right]\langle \hat{n}\rangle^{-2}(\Delta\tau)^2 \tag{129}$$

in the special case in which no scattered photons are excited initially, i.e., $\langle \hat{m} \rangle = \langle \hat{m}^2 \rangle = 0$. For the initially coherent Stokes and laser modes the factorial moment (128) reduces to the simple form $\gamma_L^{(2)} = |\alpha_S|^2 (\Delta\tau)^2$.

Our short-time solutions for the Stokes mode are

$$\begin{aligned}
\langle \hat{m}(\tau) \rangle &= \langle \hat{m} \rangle + \langle \hat{n} \rangle (\langle \hat{m} \rangle + 1)\, \Delta\tau \\
&\quad - \left[\langle \hat{m}^2 \rangle \langle \hat{n} \rangle + \langle \hat{m} \rangle (3\langle \hat{n} \rangle - \langle \hat{n}^2 \rangle) + 2\langle \hat{n} \rangle - \langle \hat{n}^2 \rangle \right] \frac{(\Delta\tau)^2}{2} \\
&\quad + \left[\langle \hat{m}^3 \rangle \langle \hat{n} \rangle + \langle \hat{m}^2 \rangle (7\langle \hat{n} \rangle - 4\langle \hat{n}^2 \rangle) + \langle \hat{m} \rangle (14\langle \hat{n} \rangle \right. \\
&\qquad \left. - 12\langle \hat{n}^2 \rangle + \langle \hat{n}^3 \rangle) + 8\langle \hat{n} \rangle - 8\langle \hat{n}^2 \rangle + \langle \hat{n}^3 \rangle \right] \frac{(\Delta\tau)^3}{6}
\end{aligned} \tag{130}$$

$$\begin{aligned}
\langle \hat{m}^2(\tau) \rangle &= \langle \hat{m}^2 \rangle + \langle \hat{n} \rangle (2\langle \hat{m}^2 \rangle + 3\langle \hat{m} \rangle + 1)\, \Delta\tau \\
&\quad - \left[2\langle \hat{m}^3 \rangle \langle \hat{n} \rangle + \langle \hat{m}^2 \rangle (9\langle \hat{n} \rangle - 4\langle \hat{n}^2 \rangle) \right. \\
&\qquad \left. + \langle \hat{m} \rangle (13\langle \hat{n} \rangle - 9\langle \hat{n}^2 \rangle) + 6\langle \hat{n} \rangle - 5\langle \hat{n}^2 \rangle \right] \frac{(\Delta\tau)^2}{2} \\
&\quad + \left[2\langle \hat{m}^4 \rangle \langle \hat{n} \rangle + \langle \hat{m}^3 \rangle (21\langle \hat{n} \rangle - 14\langle \hat{n}^2 \rangle) \right. \\
&\qquad + \langle \hat{m}^2 \rangle (73\langle \hat{n} \rangle - 72\langle \hat{n}^2 \rangle + 8\langle \hat{n}^3 \rangle) \\
&\qquad + \langle \hat{m} \rangle (102\langle \hat{n} \rangle - 118\langle \hat{n}^2 \rangle + 21\langle \hat{n}^3 \rangle) \\
&\qquad \left. + 48\langle \hat{n} \rangle - 60\langle \hat{n}^2 \rangle + 13\langle \hat{n}^3 \rangle \right] \frac{(\Delta\tau)^3}{6}
\end{aligned} \tag{131}$$

On adding Eqs. (126) and (130) we note that the sum of the mean number of photons in both the Stokes and laser modes is constant (at least up to $(\Delta\tau)^2$):

$$\langle \hat{n}(\tau) \rangle + \langle \hat{m}(\tau) \rangle = \langle \hat{n} \rangle + \langle \hat{m} \rangle \tag{132}$$

Taking a closer look at Eq. (125), which contains only terms with ρ_{nm} and $\rho_{n+1,\, m-1}$, one can draw the more fundamental conclusion that the property (132) holds for any times, in particular for the steady solutions for $\tau \to \infty$. Equation (132) is a special case of (62). Actually, we note in view of the master equation (123) that the operator $\hat{a}_L^+(\tau)\hat{a}_L(\tau) + \hat{a}_S^+(\tau)\hat{a}_S(\tau)$ is a constant of motion.

The time evolution of the mean values $\langle \hat{n}(\tau) \rangle$, $\langle \hat{m}(\tau) \rangle$, $\langle \hat{n}^2(\tau) \rangle$, and $\langle \hat{m}^2(\tau) \rangle$ is shown in Figs. 2 and 3 for initially coherent distributions. Curves B are obtained from Eqs. (126), (127), (130), and (131).

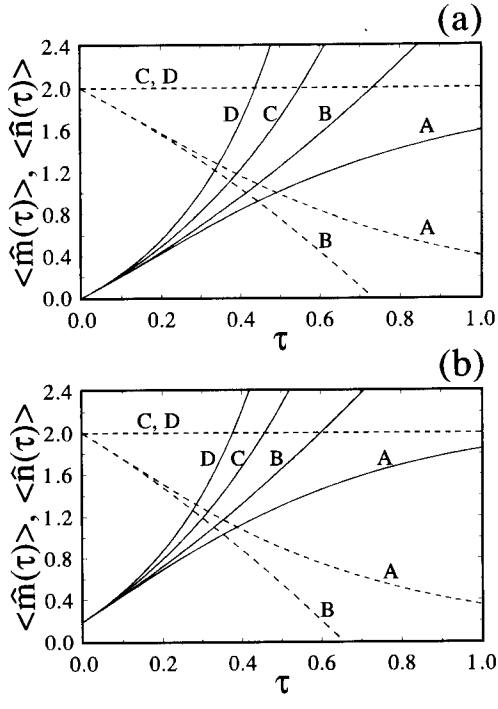

Figure 2. Time behavior of the mean number of the Stokes photons $\langle\hat{m}(t)\rangle$ (solid lines) and the laser photons $\langle\hat{n}(t)\rangle$ (dashed lines) for the initial fields: (a) $|\alpha_L = \sqrt{2}\rangle$, $|\alpha_S = 0\rangle$, and (b) $|\alpha_L = \sqrt{2}\rangle$, $|\alpha_S = \sqrt{0.2}\rangle$. Numerical results with exact solutions of Section VI.A.2 (curves A); short-time approximation of Section VI.A.1 (curves B); parametric approximation of Section V.B (curves C); approximate solutions of Section VI.B (curves D).

The factorial moment $\gamma_S^{(2)}(\tau)$, in the case of nonzero $\langle\hat{m}\rangle$, is equal to

$$\begin{aligned}\gamma_S^{(2)}(\tau) = \gamma_S^{(2)} &- 2\big(\langle\hat{m}^2\rangle - 2\langle\hat{m}\rangle^2 - \langle\hat{m}\rangle\big)\langle\hat{n}\rangle\langle\hat{m}\rangle^{-3}\Delta\tau \\ &- \Big[\langle\hat{m}^3\rangle\langle\hat{m}\rangle^2\langle\hat{n}\rangle - \langle\hat{m}^2\rangle^2\langle\hat{m}\rangle\langle\hat{n}\rangle \\ &+ \langle\hat{m}^2\rangle\langle\hat{m}\rangle^2\big(\langle\hat{n}\rangle^2 + 2\langle\hat{n}\rangle - \langle\hat{n}^2\rangle\big) \\ &- \langle\hat{m}^2\rangle\langle\hat{m}\rangle\big(2\langle\hat{n}\rangle^2 + 2\langle\hat{n}\rangle - \langle\hat{n}^2\rangle\big) \\ &- 3\langle\hat{m}^2\rangle\langle\hat{n}\rangle^2 + \langle\hat{m}\rangle^3\big(7\langle\hat{n}\rangle^2 + 8\langle\hat{n}\rangle - 5\langle\hat{n}^2\rangle\big) \\ &+ \langle\hat{m}\rangle^2\big(10\langle\hat{n}\rangle^2 + 4\langle\hat{n}\rangle - 3\langle\hat{n}^2\rangle\big) \\ &+ 3\langle\hat{m}\rangle\langle\hat{n}\rangle^2\Big]\langle\hat{m}\rangle^{-4}(\Delta\tau)^2\end{aligned} \tag{133}$$

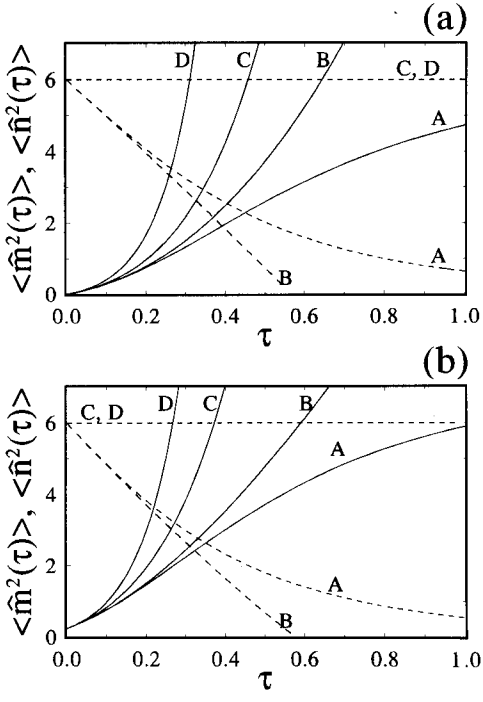

Figure 3. Time behavior of the mean-square number of the Stokes photons $\langle\hat{m}^2(t)\rangle$ (solid lines) and the laser photons $\langle\hat{n}^2(t)\rangle$ (dashed lines) for the same cases as in Fig. 2.

whereas in the case when all moments $\langle\hat{m}^k\rangle$ (for $k = 1, 2, \ldots$) are zero, $\gamma_S^{(2)}(\tau)$ can be expressed as

$$\gamma_S^{(2)}(\tau) = 2\gamma_L^{(2)} + 1 + \left(6\langle\hat{n}^3\rangle - \frac{6\langle\hat{n}^2\rangle^2}{\langle\hat{n}\rangle} - 8\langle\hat{n}^2\rangle + 8\langle\hat{n}\rangle\right)\langle\hat{n}\rangle^{-2}\frac{\Delta\tau}{3} \tag{134}$$

To obtain a correct time dependence of the factorial moment (134), it is clearly necessary to include in Eqs. (130) and (131) terms at least up to third order in τ. An equation similar to (134) has been obtained by Simaan.[75] In Fig. 4 we compare, in particular, our result for the factorial moments of photon number in the Stokes mode calculated with Eq. (133) and (134) (curves B) with that obtained from the exact solution (curves A) of the master equation (125) discussed in Section VI.A.2. Our Eq. (134) gives much better approximation to the exact results than Simaan's

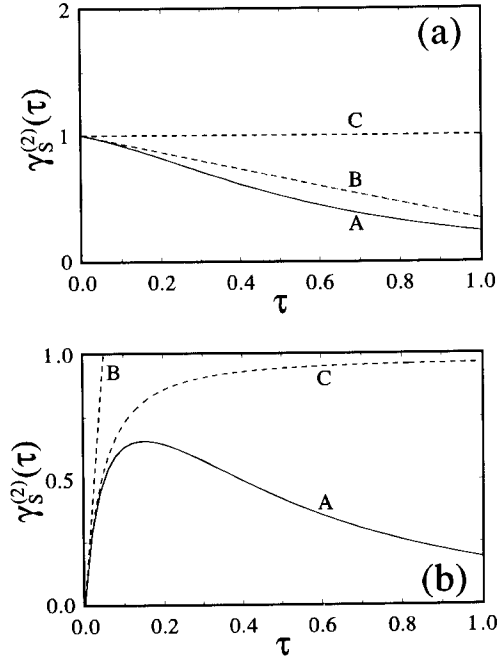

Figure 4. Time behavior of the normalized factorial moments $\gamma_S^{(2)}(\tau)$ for the Stokes mode for the same cases (except for curve D) as in Fig. 2.

formula (33) in Ref. 75. Analogously in Fig. 5, the factorial moments for the laser mode calculated with Eqs. (128) and (129) (curves B) are compared, in particular, with the exact solutions (curves A).

The Eqs. (133) and (134) reduce, respectively, to

$$\gamma_S^{(2)}(\tau) = 2|\alpha_L|^2|\alpha_S|^{-2}\Delta\tau - \left(2|\alpha_S|^4 + 3|\alpha_S|^2 + 3|\alpha_L|^2 + |\alpha_S|^2|\alpha_L|^2\right)|\alpha_L|^2|\alpha_S|^{-4}(\Delta\tau)^2 \tag{135}$$

$$\gamma_S^{(2)}(\tau) = 1 - \tfrac{2}{3}\Delta\tau \tag{136}$$

for initially coherent radiation fields.

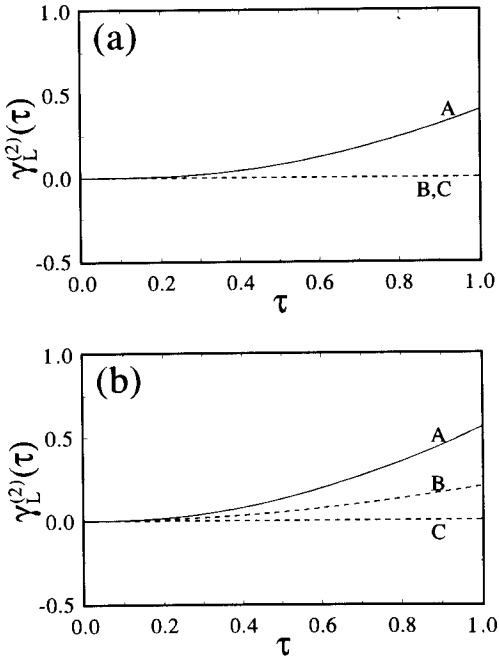

Figure 5. Same as Fig. 4, but for the normalized factorial moments $\gamma_L^{(2)}(\tau)$ of the laser mode.

The corresponding short-time dependence of the cross-correlation (interbeam) function is

$$\begin{aligned}\langle \hat{n}(\tau)\hat{m}(\tau)\rangle = \langle \hat{n}\rangle\langle \hat{m}\rangle &+ \left[\langle \hat{n}^2\rangle(\langle \hat{m}\rangle + 1) - \langle \hat{n}\rangle(\langle \hat{m}^2\rangle + 2\langle \hat{m}\rangle + 1)\right]\Delta\tau \\ &+ \Big[\langle \hat{n}^3\rangle(\langle \hat{m}\rangle + 1) - \langle \hat{n}^2\rangle(4\langle \hat{m}^2\rangle + 11\langle \hat{m}\rangle + 7) \\ &\quad + \langle \hat{n}\rangle(\langle \hat{m}^3\rangle + 6\langle \hat{m}^2\rangle + 11\langle \hat{m}\rangle + 6)\Big]\frac{(\Delta\tau)^2}{2} \\ &- \Big[3\langle \hat{n}^4\rangle(2\langle \hat{m}\rangle + \langle \hat{m}^2\rangle + 1) - \langle \hat{n}^3\rangle(3\langle \hat{m}\rangle + 4\langle \hat{m}^2\rangle - 1) \\ &\quad - \langle \hat{n}^2\rangle(68\langle \hat{m}\rangle + 43\langle \hat{m}^2\rangle + 11\langle \hat{m}^3\rangle + 36) \\ &+ \langle \hat{n}\rangle(66\langle \hat{m}\rangle + 47\langle \hat{m}^2\rangle + 14\langle \hat{m}^3\rangle + \langle \hat{m}^4\rangle + 32)\Big]\frac{(\Delta\tau)^3}{6}\end{aligned} \tag{137}$$

On inserting (126), (130), and (137) into the definition (38) of the inter-beam degree of second-order coherence, $g_{LS}^{(2)}(\tau)$, we obtain the following relation for the case when photons are initially present in the Stokes mode:

$$\begin{aligned} g_{LS}^{(2)}(\tau) = & \Big[\langle\hat{n}^2\rangle(\langle\hat{m}\rangle + 1) + \langle\hat{n}\rangle\big(\langle\hat{m}\rangle^2 - \langle\hat{m}\rangle - \langle\hat{m}^2\rangle - 1\big) \\ & - \langle\hat{n}\rangle^2(\langle\hat{m}\rangle + 1)\Big]\big(\langle\hat{n}\rangle\langle\hat{m}\rangle\big)^{-1}\Delta\tau \\ & + \Big\{\langle\hat{n}^3\rangle\langle\hat{m}\rangle(\langle\hat{m}\rangle + 1) + \langle\hat{n}^2\rangle\langle\hat{m}\rangle \\ & \times\big(3\langle\hat{m}\rangle^2 - 6\langle\hat{m}\rangle - 4\langle\hat{m}^2\rangle - 5\big) \\ & - \langle\hat{n}^2\rangle\langle\hat{n}\rangle\big(3\langle\hat{m}\rangle^2 + 5\langle\hat{m}\rangle + 2\big) \\ & + \langle\hat{n}\rangle\langle\hat{m}\rangle\Big[2\langle\hat{m}\rangle^3 - 3\langle\hat{m}\rangle^2 + \langle\hat{m}\rangle(5 - 3\langle\hat{m}^2\rangle) \\ & + 4\langle\hat{m}^2\rangle + \langle\hat{m}^3\rangle + 4\Big] \\ & - \langle\hat{n}\rangle^2\Big[2\langle\hat{m}\rangle^3 - 3\langle\hat{m}\rangle^2 - 3\langle\hat{m}\rangle(\langle\hat{m}^2\rangle + 2) - 2\big(\langle\hat{m}^2\rangle + 1\big)\Big] \\ & + 2\langle\hat{n}\rangle^3\big(\langle\hat{m}\rangle^2 + 2\langle\hat{m}\rangle + 1\big)\Big\}\langle\hat{m}\rangle^{-2}\langle\hat{n}\rangle\frac{(\Delta\tau)^2}{2} \end{aligned} \tag{138}$$

Otherwise, for the case $\langle\hat{m}\rangle = \langle\hat{m}^2\rangle = \langle\hat{m}^3\rangle = \langle\hat{m}^4\rangle = 0$, we get

$$\begin{aligned} g_{LS}^{(2)}(\tau) = & \gamma_L^{(2)} + \big(\langle\hat{n}^3\rangle - \langle\hat{n}^2\rangle^2/\langle\hat{n}\rangle \\ & - 2\langle\hat{n}^2\rangle + 2\langle\hat{n}\rangle\big)\langle\hat{n}\rangle^{-2}\frac{\Delta\tau}{2} \\ & - \big(6\langle\hat{n}^4\rangle\langle\hat{n}\rangle^2 + 5\langle\hat{n}^3\rangle\langle\hat{n}^2\rangle\langle\hat{n}\rangle \\ & - 12\langle\hat{n}^3\rangle\langle\hat{n}\rangle^2 - 3\langle\hat{n}^2\rangle^3 - 22\langle\hat{n}^2\rangle^2\langle\hat{n}\rangle \\ & + 26\langle\hat{n}^2\rangle\langle\hat{n}\rangle^2\big)\langle\hat{n}\rangle^{-4}\frac{(\Delta\tau)^2}{12} \end{aligned} \tag{139}$$

Assuming that the Stokes and laser modes are initially coherent, Eqs.

(138) and (139) reduce respectively to

$$\gamma_{LS}^{(2)}(\tau) = -\Delta\tau + \left(|\alpha_S|^2 - 2|\alpha_S|^2|\alpha_L|^2 + |\alpha_L|^2\right)|\alpha_S|^{-2}\frac{(\Delta\tau)^2}{2} \quad (140)$$

$$\gamma_{LS}^{(2)}(\tau) = -\frac{\Delta\tau}{2} + \left(1 - 13|\alpha_L|^2 - 8|\alpha_L|^4\right)\frac{(\Delta\tau)^2}{12} \quad (141)$$

Equation (137), calculated up to the third order in $\Delta\tau$, enables us to determine the relation (139) correct up to $\Delta\tau$ squared only. Simaan[75] has calculated an expression similar to Eq. (139). Examples of the time evolution of $g_{LS}^{(2)}(\tau)$ for initially coherent fields are presented in Fig. 6. Curves B in Figs. 6a and b are calculated with Eqs. (139) and (138) (including terms up to $\Delta\tau$ only). Curve S in Fig. 6a is calculated from the Simaan short-time approximate solution (32) of Ref. 75. One can compare these results (curves B and S) with $g_{LS}^{(2)}(\tau)$ obtained from our numerical calculations utilizing the exact solution of the master equation (123) (curves A). We note the supremacy of our short-time approximation (141).

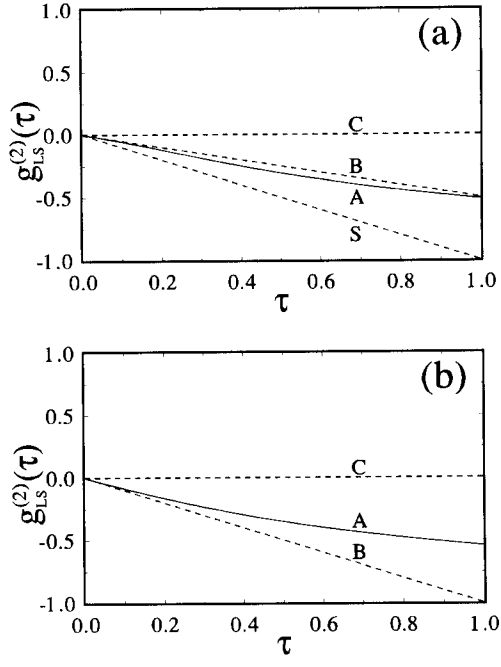

Figure 6. Same as Fig. 4, but for the interbeam degree of coherence $g_{LS}^{(2)}(\tau)$. Additional curve S is calculated with the Simaan short-time approximation (Eq. (32) of Ref. 75).

By analogy to the photon-number moments we calculate, in the short-time approximation, the mean and mean square of the annihilation operators, $\langle \hat{a}_k^+(\tau)\rangle$ and $\langle \hat{a}_k^{+2}(\tau)\rangle$ for both fields ($k = L, S$), as well as the cross-correlation functions $\langle \hat{a}_L^+(\tau)\hat{a}_S^+(\tau)\rangle$ and $\langle \hat{a}_L(\tau)\hat{a}_S^+(\tau)\rangle$. After some algebra, we arrive at

$$\langle \hat{a}_S^+(\tau)\rangle = \langle \hat{a}_S^+\rangle + \langle \hat{a}_L^+\hat{a}_L\rangle\langle \hat{a}_S^+\rangle \frac{\Delta\tau}{2} + \left(\langle \hat{a}_L^{+2}\hat{a}_L^2\rangle\langle \hat{a}_S^+\rangle - 2\langle \hat{a}_L^+\hat{a}_L\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle - 3\langle \hat{a}_L^+\hat{a}_L\rangle\langle \hat{a}_S^+\rangle\right)\frac{(\Delta\tau)^2}{8} \tag{142}$$

$$\langle \hat{a}_L^+(\tau)\rangle = \langle \hat{a}_L^+\rangle - \langle \hat{a}_L^+\rangle\left(\langle \hat{a}_S^+\hat{a}_S\rangle + 1\right)\frac{\Delta\tau}{2} + \left[\langle \hat{a}_L^+\rangle\langle \hat{a}_S^{+2}\hat{a}_S^2\rangle - 2\langle \hat{a}_L^{+2}\hat{a}_L\rangle\left(\langle \hat{a}_S^+\hat{a}_S\rangle + 1\right) + \langle \hat{a}_L^+\rangle\left(3\langle \hat{a}_S^+\hat{a}_S\rangle + 1\right)\right]\frac{(\Delta\tau)^2}{8} \tag{143}$$

$$\langle \hat{a}_S^{+2}(\tau)\rangle = \langle \hat{a}_S^{+2}\rangle + \langle \hat{a}_L^+\hat{a}_L\rangle\langle \hat{a}_S^{+2}\rangle\,\Delta\tau + \left(\langle \hat{a}_L^{+2}\hat{a}_L^2\rangle\langle \hat{a}_S^{+2}\rangle - \langle \hat{a}_L^+\hat{a}_L\rangle\langle \hat{a}_S^{+3}\hat{a}_S\rangle - 2\langle \hat{a}_L^+\hat{a}_L\rangle\langle \hat{a}_S^{+2}\rangle\right)\frac{(\Delta\tau)^2}{2} \tag{144}$$

$$\langle \hat{a}_L^{+2}(\tau)\rangle = \langle \hat{a}_L^{+2}\rangle - \langle \hat{a}_L^{+2}\rangle\left(\langle \hat{a}_S^+\hat{a}_S\rangle + 1\right)\Delta\tau + \left[\langle \hat{a}_L^{+2}\rangle\langle \hat{a}_S^{+2}\hat{a}_S^2\rangle - \langle \hat{a}_L^{+3}\hat{a}_L\rangle\left(\langle \hat{a}_S^+\hat{a}_S\rangle + 1\right) + \langle \hat{a}_L^{+2}\rangle\left(3\langle \hat{a}_S^+\hat{a}_S\rangle + 1\right)\right]\frac{(\Delta\tau)^2}{2} \tag{145}$$

$$\begin{aligned}\langle \hat{a}_L^+(\tau)\hat{a}_S^+(\tau)\rangle &= \langle \hat{a}_L^+\rangle\langle \hat{a}_S^+\rangle + \left(\langle \hat{a}_L^{+2}\hat{a}_L\rangle\langle \hat{a}_S^+\rangle - \langle \hat{a}_L^+\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle - 2\langle \hat{a}_L^+\rangle\langle \hat{a}_S^+\rangle\right)\frac{\Delta\tau}{2} \\ &\quad + \left(\langle \hat{a}_L^{+3}\hat{a}_L^2\rangle\langle \hat{a}_S^+\rangle - 11\langle \hat{a}_L^{+2}\hat{a}_L\rangle\langle \hat{a}_S^+\rangle - 6\langle \hat{a}_L^{+2}\hat{a}_L\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle\right. \\ &\quad \left. + 5\langle \hat{a}_L^+\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle + \langle \hat{a}_L^+\rangle\langle \hat{a}_S^{+3}\hat{a}_S^2\rangle + 4\langle \hat{a}_L^+\rangle\langle \hat{a}_S^+\rangle\right)\frac{(\Delta\tau)^2}{8}\end{aligned} \tag{146}$$

$$\begin{aligned}\langle \hat{a}_L(\tau)\hat{a}_S^+(\tau)\rangle &= \langle \hat{a}_L\rangle\langle \hat{a}_S^+\rangle + \left(\langle \hat{a}_L^+\hat{a}_L^2\rangle\langle \hat{a}_S^+\rangle - \langle \hat{a}_L\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle - 2\langle \hat{a}_L\rangle\langle \hat{a}_S^+\rangle\right)\frac{\Delta\tau}{2} \\ &\quad + \left(\langle \hat{a}_L^{+2}\hat{a}_L^3\rangle\langle \hat{a}_S^+\rangle - 11\langle \hat{a}_L^+\hat{a}_L^2\rangle\langle \hat{a}_S^+\rangle - 6\langle \hat{a}_L^+\hat{a}_L^2\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle\right. \\ &\quad \left. + 9\langle \hat{a}_L\rangle\langle \hat{a}_S^{+2}\hat{a}_S\rangle + \langle \hat{a}_L\rangle\langle \hat{a}_S^{+3}\hat{a}_S^2\rangle + 12\langle \hat{a}_L\rangle\langle \hat{a}_S^+\rangle\right)\frac{(\Delta\tau)^2}{8}\end{aligned} \tag{147}$$

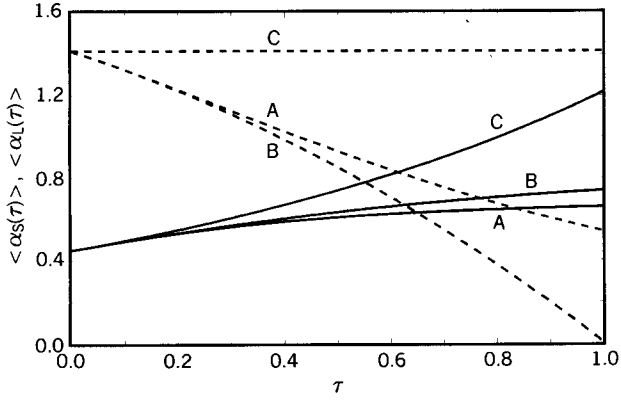

Figure 7. Time dependence of the expectation values of the field amplitudes $\langle \hat{a}_S(\tau)\rangle = \langle \alpha_S(\tau)\rangle$ (solid lines) and $\langle \hat{a}_L(\tau)\rangle = \langle \alpha_L(\tau)\rangle$ (dashed lines) for fields initially coherent $|\alpha_L = \sqrt{2}\rangle$ and $|\alpha_S = \sqrt{0.2}\rangle$. Curves A, B, C are calculated within the formalisms of Sections VI.A.2, VI.A.1, and V.B, respectively.

For brevity, here, we shall restrict our considerations to initially coherent states for the Stokes mode denoted as $\alpha_S = |\alpha_S| \exp(i\phi_S)$ and for the laser mode $\alpha_L = |\alpha_L| \exp(i\phi_L)$. In Figs. 7 and 8 we demonstrate the evolution of our short-time approximations for $\langle \hat{a}_S(\tau)\rangle$ (solid line B in Fig. 7), $\langle \hat{a}_L(\tau)\rangle$ (dashed line B in Fig. 7), $\langle \hat{a}_S^2(\tau)\rangle$ (solid line B in Fig. 8), and $\langle \hat{a}_L^2(\tau)\rangle$ (dashed line B in Fig. 8) for initially coherent radiation modes.

From the general relations (130) and (126), under the condition of initially coherent Stokes and laser fields, we get

$$\langle \hat{m}(\tau)\rangle = |\alpha_S|^2 + |\alpha_L|^2\big(|\alpha_S|^2 + 1\big)\,\Delta\tau + |\alpha_L|^2\Big[\big(|\alpha_L|^2 + 1\big)\big(|\alpha_S|^2 + 1\big) - \big(|\alpha_S|^4 + 4|\alpha_S|^2 + 2\big)\Big]\frac{(\Delta\tau)^2}{2} \tag{148}$$

$$\langle \hat{n}(\tau)\rangle = |\alpha_L|^2 + |\alpha_S|^2 - \langle \hat{m}(\tau)\rangle \tag{149}$$

Inserting (148) as well as (142) and (144) with $\langle \hat{a}_k^{+p}\hat{a}_k^q\rangle = |\alpha_k|^{p+q}\exp[-i(p-q)\phi_k]$ $(k = S, A)$, into (43) we obtain the θ-dependent variance for the Stokes mode:

$$\Big\langle \big(\Delta\hat{X}_S(\theta)\big)^2\Big\rangle = 1 + 2|\alpha_L|^2\Delta\tau + |\alpha_L|^2\Big\{|\alpha_L|^2 - \big[1 + \cos^2(\theta - \phi_S)\big]|\alpha_S|^2 - 1\Big\}(\Delta\tau)^2. \tag{150}$$

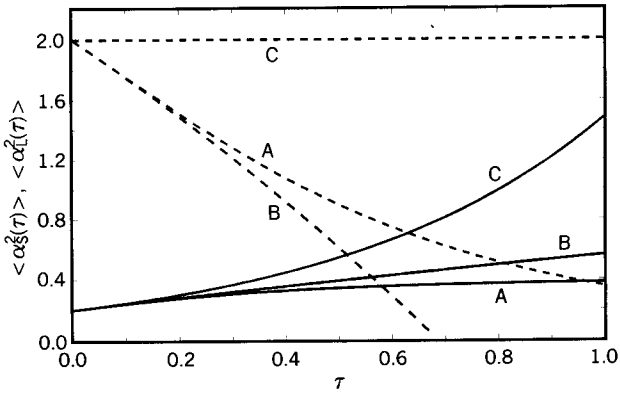

Figure 8. Time dependence of $\langle\hat{a}_S^2(\tau)\rangle = \langle\alpha_S^2(\tau)\rangle$ (solid lines) and $\langle\hat{a}_L^2(\tau)\rangle = \langle\alpha_L^2(\tau)\rangle$ (dashed lines) for the same cases as in Fig. 7.

The minimal variance $\langle(\Delta\hat{X}_{S-})^2\rangle$, which follows from (46) or directly from (150), is equal to

$$\left\langle\left(\Delta\hat{X}_{S-}\right)^2\right\rangle = 1 + 2|\alpha_L|^2\Delta\tau + |\alpha_L|^2\left(|\alpha_L|^2 - 2|\alpha_S|^2 - 1\right)(\Delta\tau)^2 \quad (151)$$

Analogously, for the laser field we obtain the following θ-dependent variance:

$$\left\langle\left(\Delta\hat{X}_L(\theta)\right)^2\right\rangle = 1 + \tfrac{1}{2}\left[\cos(2\theta - 2\phi_L) + 1\right]|\alpha_L|^2|\alpha_S|^2(\Delta\tau)^2 \quad (152)$$

on insertion of Eqs. (149), (143), and (145) into (43). With regard to the relation (46), the minimal variance for the field, $\langle(\Delta\hat{X}_{L-})^2\rangle$, is constant up to the second order in time:

$$\left\langle\left(\Delta\hat{X}_{L-}\right)^2\right\rangle = 1 \quad (153)$$

One can readily deduce the maximal variances $\langle(\Delta\hat{X}_{L,S\pm})^2\rangle$ from (150) and (152) or from (46). The time evolution of the single-mode extremal variances obtained from (150)–(153) is presented in Figs. 9 and 10 (for $\phi_L = 0$): $\langle(\Delta\hat{X}_{S-}(\tau))^2\rangle = \langle(\Delta\hat{X}_{S2}(\tau))^2\rangle$ (solid line B in Fig. 9), $\langle(\Delta\hat{X}_{S+}(\tau))^2\rangle = \langle(\Delta\hat{X}_{S1}(\tau))^2\rangle$ (dashed line B in Fig. 9), $\langle(\Delta\hat{X}_{L-}(\tau))^2\rangle = \langle(\Delta\hat{X}_{L1}(\tau))^2\rangle$ (solid line B in Fig. 10), $\langle(\Delta\hat{X}_{L+}(\tau))^2\rangle = \langle(\Delta\hat{X}_{L2}(\tau))^2\rangle$ (dashed line B in Fig. 10).

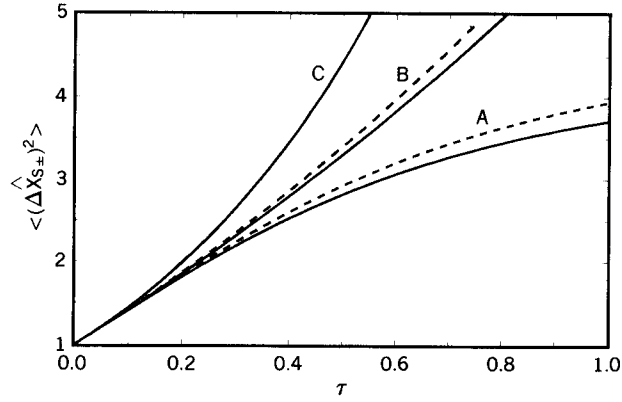
5
4
3
2
1
0.0
0.2
0.4
0.6
0.8
1.0
τ
C
B
A
<(ΔX̂S±)²>

Figure 9. Time dependence of the extremal variances $\langle(\Delta\hat{X}_{S-})^2\rangle$ (solid lines) and $\langle(\Delta\hat{X}_{S+})^2\rangle$ (dashed lines) for the same cases as in Fig. 7.

The covariances for quadratures in the Stokes and laser mode, according to (52), are, respectively,

$$\left\langle\left\{\Delta\hat{X}_{S1}, \Delta\hat{X}_{S2}\right\}\right\rangle = |\alpha_L|^2|\alpha_S|^2 \sin(2\phi_S)(\Delta\tau)^2 \tag{154}$$

$$\left\langle\left\{\Delta\hat{X}_{L1}, \Delta\hat{X}_{L2}\right\}\right\rangle = -|\alpha_L|^2|\alpha_S|^2 \sin(2\phi_L)(\Delta\tau)^2 \tag{155}$$

The generalized Heisenberg uncertainty relation (51) with the covariances (154) and (155) inserted takes the following form for the Stokes mode in

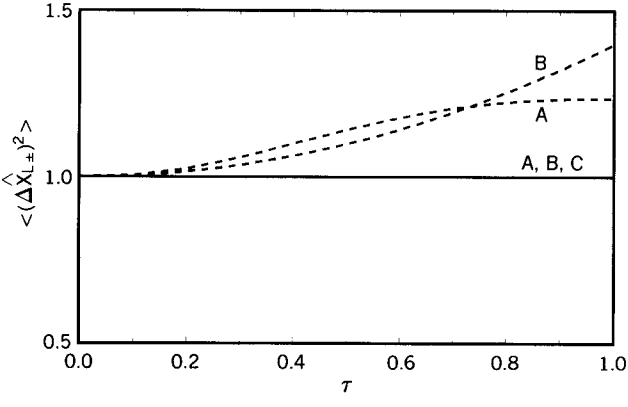
1.5
1.0
0.5
0.0
0.2
0.4
0.6
0.8
1.0
τ
B
A
A, B, C
<(ΔX̂L±)²>

Figure 10. Time dependence of the extremal variances $\langle(\Delta\hat{X}_{L-})^2\rangle$ (solid lines) and $\langle(\Delta\hat{X}_{L+})^2\rangle$ (dashed lines) for the same cases as in Fig. 7.

our short-time approximation:

$$4|\alpha_L|^2\Delta\tau + |\alpha_L|^2\big(6|\alpha_L|^2 - 3|\alpha_S|^2 - 2\big)(\Delta\tau)^2 \geq 0 \tag{156}$$

and for the laser mode

$$2|\alpha_L|^2|\alpha_S|^2(\Delta\tau)^2 \geq 0 \tag{157}$$

To obtain the two-mode variances and covariances of the quadratures one has to calculate, apart from the single-mode functions (150), (152), (154), and (155), the cross-correlations (57), which are obtained in the following form:

$$\begin{aligned}\langle \Delta\hat{X}_{L1}\,\Delta\hat{X}_{S1}\rangle &= -|\alpha_L|\,|\alpha_S|\Big\{2\cos\phi_L\cos\phi_S\,\Delta\tau + \big[\cos(\phi_L-\phi_S)(4n_L - 6n_S - 11)\\ &\quad + \cos(\phi_L+\phi_S)\big(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\big)\big]\frac{(\Delta\tau)^2}{4}\Big\}\end{aligned} \tag{158}$$

$$\begin{aligned}\langle \Delta\hat{X}_{L2}\,\Delta\hat{X}_{S2}\rangle &= -|\alpha_L|\,|\alpha_S|\Big\{2\sin\phi_L\sin\phi_S\,\Delta\tau + \big[\cos(\phi_L-\phi_S)(4n_L - 6n_S - 11)\\ &\quad - \cos(\phi_L+\phi_S)\big(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\big)\big]\frac{(\Delta\tau)^2}{4}\Big\}\end{aligned} \tag{159}$$

$$\begin{aligned}\langle \Delta\hat{X}_{L1}\,\Delta\hat{X}_{S2}\rangle &= |\alpha_L|\,|\alpha_S|\Big\{2\cos\phi_L\sin\phi_S\,\Delta\tau + \big[\sin(\phi_S-\phi_L)(4n_L - 6n_S - 11)\\ &\quad + \sin(\phi_S+\phi_L)\big(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\big)\big]\frac{(\Delta\tau)^2}{4}\Big\}\end{aligned} \tag{160}$$

$$\begin{aligned}\langle \Delta\hat{X}_{L2}\,\Delta\hat{X}_{S1}\rangle &= |\alpha_L|\,|\alpha_S|\Big\{2\sin\phi_L\cos\phi_S\,\Delta\tau + \big[\sin(\phi_L-\phi_S)(4n_L - 6n_S - 11)\\ &\quad + \sin(\phi_L+\phi_S)\big(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\big)\big]\frac{(\Delta\tau)^2}{4}\Big\}\end{aligned} \tag{161}$$

Thus, the two-mode Wigner covariance (56) of the quadratures $\hat{X}_{LS1}$ and $\hat{X}_{LS2}$ is

$$\begin{aligned}\left\langle\left\{\Delta\hat{X}_{LS1}, \Delta\hat{X}_{LS2}\right\}\right\rangle &= 4|\alpha_L|\,|\alpha_S|\sin(\phi_L+\phi_S)\,\Delta\tau \\ &\quad + \left[4|\alpha_L|^2|\alpha_S|^2\cos\tfrac{1}{2}(\phi_S+\phi_L)\sin\tfrac{1}{2}(\phi_S-\phi_L)\right. \\ &\quad\quad + |\alpha_L|\,|\alpha_S|\sin(\phi_L+\phi_S) \\ &\quad\quad \left.\times\left(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\right)\right](\Delta\tau)^2 \qquad (162)\end{aligned}$$

The two-mode variances (55) of $\hat{X}_{LS1}$ and $\hat{X}_{LS2}$ are

$$\begin{aligned}&\left\langle\left(\Delta\hat{X}_{LS1,2}\right)^2\right\rangle \\ &\quad = 2 + 2\left\{|\alpha_L|^2 - |\alpha_L|\,\alpha_S|\left[\cos(\phi_L-\phi_S) \pm \cos(\phi_L+\phi_S)\right]\right\}\Delta\tau \\ &\quad\quad + \left\{2|\alpha_L|^2\left(|\alpha_L|^2 - |\alpha_S|^2 - 1\right) \pm |\alpha_L|^2|\alpha_S|^2(\cos 2\phi_L - \cos 2\phi_S)\right. \\ &\quad\quad\quad - |\alpha_S|\,|\alpha_L|\left[\cos(\phi_L-\phi_S)\left(4|\alpha_L|^2 - 6|\alpha_S|^2 - 11\right)\right. \qquad (163) \\ &\quad\quad\quad\quad \left.\left.\pm\cos(\phi_L+\phi_S)\left(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\right)\right]\right\}\frac{(\Delta\tau)^2}{2}\end{aligned}$$

whereas the extremal variances are

$$\begin{aligned}\left\langle\left(\Delta\hat{X}_{LS\pm}\right)^2\right\rangle &= 2 + 2\left[|\alpha_L|^2 - |\alpha_L|\,|\alpha_S|\cos(\phi_L-\phi_S)\right]\Delta\tau \\ &\quad + \left[|\alpha_L|^2\left(|\alpha_L|^2 - |\alpha_S|^2 - 1\right)\right. \\ &\quad\quad \left. - |\alpha_L|\,|\alpha_S|\cos(\phi_L-\phi_S)\left(2|\alpha_L|^2 - 3|\alpha_S|^2 - \tfrac{11}{2}\right)\right](\Delta\tau)^2 \\ &\quad \pm\left|2\alpha_L\alpha_S\,\Delta\tau + \left[|\alpha_L|^2\alpha_S^2 - |\alpha_S|^2\alpha_L^2\right.\right. \qquad (164) \\ &\quad\quad \left.\left. + \alpha_L\alpha_S\left(4|\alpha_L|^2 - 2|\alpha_S|^2 - 3\right)\right]\frac{(\Delta\tau)^2}{2}\right|\end{aligned}$$

according to the general expression (58).

Equations (158)–(164) can be readily generalized to any initial distribution of the radiation fields.

2. *Exact Solutions*

Let us now proceed to the exact solution of the master equation (125). We apply the Laplace transform method. The method is readily applicable to nonlinear master equations for a variety of nonlinear optical phenomena (Refs. 30, 73, and 220 and references therein); in particular, it has been applied successfully to different multiphoton Raman processes in Refs. 73, 75, 78, 106, 107, 112, 115, and 116. The solution of (125) for diagonal terms of the density matrix $\rho_{nm}(00\tau)$ (i.e., for $\nu = \mu = 0$) was derived by McNeil and Walls[73] and then, in a more general form, by Simaan.[75] As the chief result of the present work we derive the time-dependence of the complete density matrix $\rho_{nm}(\nu\mu\tau)$, where the degrees of off-diagonality ν, μ are arbitrary. To the best of our knowledge, ours is the first derivation of a complete analytical solution to the Raman scattering model including depletion of the pump field.

As usual, we assume that the Stokes and laser beams are mutually independent at the initial time $\tau = \tau_0$. Thus, the initial joint distribution $\rho_{nm}(\nu\mu\tau_0)$ is a product of the distributions for the separate beams,

$$\rho_{nm}(\nu\mu\tau_0) = \rho_n^L(\nu\tau_0)\rho_m^S(\mu\tau_0) \tag{165}$$

Let us define the coefficient λ in terms of the integer-value function $[[x]]$ (the maximum integer $\leq x$):

$$\lambda = \left[\left[\frac{m-n+1}{2} + \frac{\mu-\nu}{4}\right]\right] \tag{166}$$

The exact solution of (125) under the condition (165), derived in Appendix B, reads as follows for $\lambda < 0$:

$$\begin{aligned}\rho_{nm}(\nu\mu\tau) = {} & \left[\frac{m!(m+\mu)!}{n!(n+\nu)!}\right]^{1/2} \sum_{l=0}^{m} \rho_{n+l}^L(\nu\tau_0)\rho_{m-l}^S(\mu\tau_0) \\ & \times \left[\frac{(n+l)!(n+l+\nu)!}{(m-l)!(m-l+\mu)!}\right]^{1/2} \\ & \times \sum_{q=0}^{l} \exp[-f(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q}}^{l} [f(p) - f(q)]^{-1}\end{aligned} \tag{167}$$

whereas for $\lambda \geq 0$ it is

$$\rho_{nm}(\nu\mu\tau) = \left[\frac{m!(m+\mu)!}{n!(n+\nu)!}\right]^{1/2} \times \left\{ \sum_{l=0}^{\lambda} \rho^{L}_{n+l}(\nu\tau_0)\rho^{S}_{m-l}(\mu\tau_0) \times \left[\frac{(n+l)!(n+l+\nu)!}{(m-l)!(m-l+\mu)!}\right]^{1/2} \times \sum_{q=0}^{l} \exp[-f(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q}}^{l} [f(p)-f(q)]^{-1} + (1-\delta_{m0}) \sum_{l=\lambda+1}^{m} \rho^{L}_{n+l}(\nu\tau_0)\rho^{S}_{m-l}(\mu\tau_0) \times \left[\frac{(n+l)!(n+l+\nu)!}{(m-l)!(m-l+\mu)!}\right]^{1/2} \times \sum_{q=0}^{\lambda} \sum_{q'=\lambda+1}^{l} \prod_{\substack{p=0 \\ p\neq q}}^{\lambda} [f(p)-f(q)]^{-1} \prod_{\substack{p'=\lambda+1 \\ p'\neq q'}}^{l} [f(p')-f(q')]^{-1} \times \left(\delta_{f(q)f(q')}\,\Delta\tau \exp[-f(q)\,\Delta\tau] + (\delta_{f(q)f(q')}-1) \frac{\exp[-f(q)\,\Delta\tau]-\exp[-f(q')\,\Delta\tau]}{f(q)-f(q')} \right) \right\} \tag{168}$$

where the function $f(x)$ is given by

$$f(x) = \tfrac{1}{2}[(n+x)(m-x+1) + (n+x+\nu)(m-x+\mu+1)] \tag{169}$$

The coefficient λ can be alternatively defined as

$$\lambda = \left[\left[\frac{m-n}{2} + \frac{\mu-\nu}{4}\right]\right] \tag{170}$$

The solution (167) and (168) with consequent application of the coefficient λ of Eq. (170) reduces, as it should, to the Simaan solution (45) and (49) of Ref. 75 for the diagonal matrix elements ($\nu = \mu = 0$); McNeil and Walls have also obtained a solution of (125) for the diagonal matrix elements (Eqs. (6.5), (6.6), and (4.14) in Ref. 73); however, their solution is not in

full agreement with Simaan's solution and is not a special case of ours for reasons given by Simaan.[75]

The solution (167) and (168) is very well adapted to numerical analysis; nonetheless, it is of a rather complicated form. Our solution of (125) can be rewritten more compactly. Following the method of Malakyan,[116] we find (for details, see Appendix B)

$$\begin{aligned}\rho_{nm}(\nu\mu\tau) &= \left[\frac{m!(m+\mu)!}{n!(n+\nu)!}\right]^{1/2} \sum_{l=0}^{m} \rho^{L}_{n+l}(\nu\tau_0)\rho^{S}_{m-l}(\mu\tau_0) \\ &\times \left[\frac{(n+l)!(n+l+\nu)!}{(m-l)!(m-l+\mu)!}\right]^{1/2} \\ &\times \hat{\mathscr{D}} \sum_{\substack{q=0 \\ q\neq q'_1,q'_2,\ldots,q'_d}}^{l} \exp[-f(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q,q'_1,q'_2,\ldots,q'_d}}^{l} [f(p)-f(q)]^{-1}\end{aligned} \tag{171}$$

The differential operator of the dth order, $\hat{\mathscr{D}}$, is defined as follows:

$$\hat{\mathscr{D}} = (-1)^d \prod_{r=1}^{d} \frac{\partial}{\partial f(q_r)} \tag{172}$$

The order d of the differential operator (172) is equal to the number of pairs of mutually equal factors occurring in the product of Eq. (171), $f(q_1) = f(q'_1), f(q_2) = f(q'_2), \ldots, f(q_d) = f(q'_d)$. If there are no pairs of equal factors, then the operator $\hat{\mathscr{D}}$ is defined to be unity (see Appendix B). The solutions (167), (168), and (171) represent the chief result of our paper. In Section V.B in the Raman effect model under the parametric approximation we have analyzed, in particular, the single-mode solutions for either the Stokes mode or for the anti-Stokes mode. For completeness, we give in Appendix C the solution for anti-Stokes scattering without the parametric approximation. The degree of off-diagonality μ is assumed to be nonnegative (contrary to ν); nonetheless, the time dependence of the complete density matrix $\rho_{nm}(\nu\mu\tau)$ is determined by the simple relation for the inverse matrix elements:

$$\rho^{*}_{nm}(\nu\mu\tau) = \rho_{n+\nu,m+\mu}(-\nu,-\mu,\tau) \tag{173}$$

Thus, solutions (167), (168), and/or (171) provide an entire specification for all measurable properties of the light field under consideration.

The two-mode (joint) density matrix with elements $\rho_{nm}(\nu\mu\tau)$, (167) and (168), enables the calculation of the single-mode (separate) density matrix with elements $\rho_m^S(\mu\tau)$ and $\rho_m^L(\mu\tau)$. The Stokes mode matrix elements $\rho_m^S(\mu\tau)$ can be calculated from

$$\rho_m^S(\mu\tau) = \sum_{n=0}^{\infty} \sum_{\nu=-n}^{\infty} \rho_{nm}(\nu\mu\tau) \tag{174}$$

and the laser mode matrix elements $\rho_m^L(\mu\tau)$ can be found analogously, with the exception that for terms $\rho_{nm}(\nu\mu\tau)$ with $\mu < 0$ the property (173) must be used. The already mentioned solution of McNeil and Walls[73] corresponds to the separate diagonal density matrix (174).

There is yet another manner of expressing the two-mode solutions of the master equation (125), $\rho_{nm}(00\tau)$, for any initial distributions, via the density matrix elements for the initial number states in the Stokes and laser fields:

$$\rho_{nm}(00\tau) = \sum_{n_0=0}^{\infty} \sum_{m_0=0}^{\infty} \rho_{nm}^{(n_0, m_0)}(00\tau)\rho_{n_0}^L(0\tau_0)\rho_{m_0}^S(0\tau_0) \tag{175}$$

where $\rho_{nm}^{(n_0, m_0)}(00\tau)$ is the solution (167) and (168) for $\rho_{nm}(00\tau)$, under the initial conditions that the laser field is in the number state $|n_0\rangle$ and the Stokes mode is in the number state $|m_0\rangle$. The weighting functions in (175) are arbitrary initial distributions of the laser, $\rho_n^L(0\tau_0)$, and the Stokes field, $\rho_n^S(0\tau_0)$. Here, for brevity, we restrict our considerations to diagonal terms (with $\nu = \mu = 0$). Otherwise, instead of $\rho_{nm}^{(n_0, m_0)}(00\tau)$ we would have to use the solution $\rho_{nm}^{(n_0, m_0, \nu_0, \mu_0)}(\nu\mu\tau)$ and perform two extra summations in Eq. (175) over ν, μ. McNeil and Walls[73] have presented their solution of Eq. (125) in this manner. Analogously, we can express the single-mode distributions $\rho_n^L(0\tau)$ ($\rho_m^S(0\tau)$) for arbitrary initial states using the solutions $\rho_n^{(m_0)}(\tau)$ ($\rho_m^{(n_0)}(\tau)$) for the initial photon-number states $|m_0\rangle(|n_0\rangle)$. For instance,for the Stokes mode solution we apply the formula

$$\rho_m^s(0\tau) = \sum_{m_0=0}^{\infty} \rho_n^{(n_0)}(\tau)\rho_{n_0}^L(0\tau_0) \tag{176}$$

We shall make use of this procedure for the diagonal approximate solutions (194).

Having the solutions (167), (168), or (171) available we can, at least numerically, analyze, e.g., the single- and two-mode photocount statistics

and quadrature squeezing. The expectation values in the relations describing squeezing and photocount statistics (see Section IV) are readily expressed in terms of the density matrix elements $\rho_{nm}(\nu\mu\tau)$ by way of

$$\langle \hat{n}^k(t) \rangle = \sum_{n,m=0}^{\infty} n^k \rho_{n,m}(0,0,t) \tag{177}$$

$$\langle \hat{m}^k(t) \rangle = \sum_{n,m} m^k \rho_{n,m}(0,0,t) \tag{178}$$

$$\langle \hat{a}_L^{+k}(t) \rangle = \sum_{n,m} \left[\frac{(n+k)!}{n!} \right]^{1/2} \rho_{n,m}(k,0,t) \tag{179}$$

$$\langle \hat{a}_S^{+k}(t) \rangle = \sum_{n,m} \left[\frac{(m+k)!}{m!} \right]^{1/2} \rho_{n,m}(0,k,t) \tag{180}$$

$$\langle \hat{a}_L^{+}(t)\hat{a}_S^{+}(t) \rangle = \sum_{n,m} [(n+1)(m+1)]^{1/2} \rho_{n,m}(1,1,t) \tag{181}$$

$$\langle \hat{a}_L^{+}(t)\hat{a}_S(t) \rangle = \sum_{n,m} [(n+1)m]^{1/2} \rho_{n,m}(1,-1,t) \tag{182}$$

In Section III we defined the s-parametrized quasiprobability distribution $\mathscr{W}^{(s)}(\{\alpha_k\})$ and the s-parametrized characteristic function $\mathscr{C}^{(s)}(\{\beta_k\})$ and adduced relations between them for any parameter s. Here, we deal with matrix elements in Fock basis of the density operator, $\rho_{nm}(\nu\mu\tau)$. To achieve consistency between our analysis of the Raman effect presented in this section with the analysis of section V, we shall present some relations between the functions $\mathscr{W}^{(s)}(\{\alpha_k\})$ or $\mathscr{C}^{(s)}(\{\beta_k\})$ and the density operator $\hat{\rho}(\{\hat{a}_k\})$. We restrict the general formulas for the M-mode fields to our two-mode situations, so that $(\{\alpha_k\}) = (\alpha_L, \alpha_{S,A})$. These formulas are in complete analogy with the results of Cahill and Glauber[197] for the single-mode case. By virtue of the operator $\hat{T}(\alpha_L, \alpha_{S,A})$, which is the Fourier transform of the s-parametrized displacement operator $\hat{D}^{(s)}(\beta_L, \beta_{S,A})$ (see Eq. (19)),

$$\hat{T}^{(s)}(\alpha_L, \alpha_{S,A}) = \frac{1}{\pi^2} \int \hat{D}^{(s)}(\beta_L, \beta_{S,A}) \times \exp(\alpha_L \beta_L^* + \alpha_{S,A}\beta_{S,A}^* - \text{c.c.}) \mathrm{d}^2\beta_L \, \mathrm{d}^2\beta_{S,A} \tag{183}$$

the density matrix $\hat{\rho}(\hat{a}_L, \hat{a}_{S,A})$ can be obtained from the s-parametrized

quasidistribution $\mathscr{W}^{(s)}(\alpha_L, \alpha_{S,A})$, (21),

$$\hat{\rho}(\hat{a}_L, \hat{a}_{S,A}) = \frac{1}{\pi^2}\int \mathscr{W}^{(s)}(\alpha_L, \alpha_{S,A}) \times \hat{T}^{(-s)}(\alpha_L, \alpha_{S,A})\mathrm{d}^2\alpha_L\, \mathrm{d}^2\alpha_{S,A} \tag{184}$$

The inverse relation,

$$\mathscr{W}^{(s)}(\alpha_L, \alpha_{S,A}) = \mathrm{Tr}\left[\hat{\rho}(\hat{a}_L, \hat{a}_{S,A})\hat{T}^{(s)}(\alpha_L, \alpha_{S,A})\right] \tag{185}$$

resembles expression (20) for the characteristic function $\mathscr{C}^{(s)}(\beta_L, \beta_{S,A})$, which is the average value of the displacement operator $\hat{D}^{(s)}(\alpha_L, \alpha_{S,A})$. We are interested in the relations for the matrix elements of $\hat{\rho}(\hat{a}_L, \hat{a}_{S,A})$. They immediately follow from (184) and (185):

$$\rho_{n,m}(\nu, \mu) = \frac{1}{\pi^2}\int \mathscr{W}^{(s)}(\alpha_L, \alpha_{S,A}) \times \left\langle n, m\middle|\hat{T}^{(-s)}(\alpha_L, \alpha_{S,A})\middle|n + \nu, m + \mu\right\rangle \mathrm{d}^2\alpha_L\, \mathrm{d}^2\alpha_{S,A} \tag{186}$$

$$\mathscr{W}^{(s)}(\alpha_L, \alpha_{S,A}) = \sum_{n=0}^{\infty}\sum_{m=0}^{\infty}\sum_{\nu=-n}^{\infty}\sum_{\mu=-m}^{\infty} \rho^*_{n,m}(\nu, \mu) \times \left\langle n, m\middle|\hat{T}^{(s)}(\alpha_L, \alpha_{S,A})\middle|n + \nu, m + \mu\right\rangle \tag{187}$$

The Fock matrix elements for the two-mode field,

$$\left\langle n, m\middle|\hat{T}^{(s)}(\alpha_L, \alpha_{S,A})\middle|n + \nu, m + \mu\right\rangle = \left\langle n\middle|\hat{T}^{(s)}(\alpha_L)\middle|n + \nu\right\rangle \left\langle m\middle|\hat{T}^{(s)}(\alpha_{S,A})\middle|m + \mu\right\rangle \tag{188}$$

are simply products of the two single-mode Fock matrix elements given by Cahill and Glauber[197]:

$$\left\langle n\middle|\hat{T}^{(s)}(\alpha_L)\middle|n + \nu\right\rangle = \sqrt{\frac{n!}{(n+\nu)!}}\left(\frac{2}{1-s}\right)^{\nu+1}\left(\frac{s+1}{s-1}\right)^n \times \exp\left(-\frac{2}{1-s}|\alpha_L|^2\right) L_n^{(\nu)}\left(\frac{4|\alpha_L|^2}{1-s^2}\right)(\alpha_L^*)^\nu \tag{189}$$

where $L_n^{(\nu)}(x)$ is the generalized Laguerre polynomial. The above equations show equivalency of the two apparently different formalisms we have

been dealing with: On the one side, the s-parametrized quasiprobability distribution functions $\mathscr{W}^{(s)}$ obtained within the Fokker-Planck equation formalism presented in Section V, and, on the other side, the density matrix operator formalism under discussion in this section.

Solutions (167), (168), or (171) reduce to rather simple expressions in special cases, for instance, on the one side, for long periods of time when the laser beam is totally depleted, and on the other, for an intense laser beam almost unaffected (undepleted) during the process of scattering. We now discuss these two cases.

3. *Long-time Solutions*

After a sufficiently long time, the system settles down to a steady state as a result of the total depletion of the laser pump. The steady-state solutions can be readily deduced from (167) and (168). Indeed, in the time limit ($\tau \to \infty$), the nonzero matrix elements $\rho_{nm}(\nu\mu\infty)$ must satisfy the condition for the function $f(x)$ (169) that $f(q) = 0$, which implies that $q = 0$. Hence, we have

$$\rho_{nm}(\nu\mu, \tau = \infty) = 0 \qquad \text{for } n, \nu \neq 0 \tag{190}$$

for arbitrary $m, m + \mu$ ranging from zero to infinity. All photon-number and annihilation operator moments for the laser beam vanish in the time limit

$$\begin{aligned} \langle \hat{n}^p(\infty) \rangle &= 0 \\ \langle \hat{a}_L^p(\infty) \rangle &= 0 \qquad \text{for } p > 0 \end{aligned} \tag{191}$$

which reflects the fact that the laser beam is totally depleted. The normalization condition takes the form

$$\sum_{m=0}^{\infty} \rho_{0m}(00\infty) = 1 \tag{192}$$

In the model of hyper-Raman scattering, as was shown by Malakyan,[116] there intervene in the limit $\tau \to \infty$ the nonzero density matrix elements $\rho_{0m}(0\mu\infty)$, $\rho_{1m}(0\mu\infty)$, $\rho_{0m}(1\mu\infty)$, and $\rho_{1m}(-1, \mu\infty)$. Hence, the photon-number moments and annihilation operator moments of the laser mode do not vanish, contrary to the model of Raman scattering under consideration in view of (191). If we assume that initially there are no photons in the Stokes mode, then $\rho_n^L(0\tau_0) = \rho_n^S(0\infty)$ (Ref. 75), which implies that an arbitrary photon-number moment $\langle \hat{m}^p(\infty) \rangle$ (with any p) for the Stokes

mode in the time limit is identical with the corresponding moment for the laser mode, $\langle \hat{n}^p(\tau_0)\rangle$, at the time $\tau = \tau_0$.

B. Raman Scattering Without Pump Depletion

Compact approximate solutions can be obtained from Eqs. (167) under the condition that the initial laser beam is much more intense than the Stokes beam, i.e., $\langle \hat{n}\rangle \gg \langle \hat{m}\rangle$. The depletion of the laser beam and amplification of the Stokes beam restrict the validity of this approximation to short evolution times τ ($\tau \ll 1$). This approximation implies that the density matrix elements $\rho_{nm}(\nu\mu\tau)$ for $\lambda \geq 0$ given by Eq. (168) are negligible. Moreover, we can simplify the remaining solution (167) by setting $n \approx n \pm m$. In the analysis of the phenomena described by the density $\rho_{nm}(\nu\mu\tau)$ with small degree of off-diagonality ν (such as quadrature squeezing), we can set $n \approx n + \nu$. Alternatively, in order to, for instance, investigate phase properties[129, 130, 131] (which require summation over ν ranging from zero to infinity) one might assume that the fluctuations in the laser beam are small in comparison to their mean value, i.e., $\langle \hat{n}\rangle \gg \sqrt{\langle (\Delta\hat{n})^2\rangle}$. Under these approximations the solution of (125) takes the form

$$\begin{aligned}\rho_{nm}(\nu\mu\tau) \approx\ & [m!(m+\mu)!]^{1/2} \\ & \times \sum_{l=0}^{m} \rho_n^L(\nu\tau_0)\rho_{m-l}^S(\mu\tau_0)[(m-l)!(m-l+\mu)!]^{-1/2} \\ & \times \sum_{q=0}^{l} \exp[-n(m-q+1+\mu/2)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q}}^{l} (q-p)^{-1}\end{aligned} \tag{193}$$

Applying the binomial theorem we rewrite (193) as

$$\begin{aligned}\rho_{nm}(\nu\mu\tau) \approx\ & \sum_{l=0}^{m} \left[\binom{m}{l}\binom{m+\mu}{l}\right]^{1/2} (e^{n\Delta\tau}-1)^l \\ & \times \exp[-n(m+1+\mu/2)\,\Delta\tau]\rho_n^L(\nu\tau_0)\rho_{m-l}^S(\mu\tau_0)\end{aligned} \tag{194}$$

which, for $\mu = 0$ and $\nu = 0$, goes over into Simaan's equation of Ref. 75. The density matrix (193), applied to relations (177)–(182), enables the calculation of the expectation values and variances for the Stokes mode and the laser mode; however, in the latter case, as a result of the approximations assumed, we find no time dependence of the laser field photon-number moments for a Stokes beam initially in a number state

containing

$$\langle f[\hat{n}(\tau)]\rangle = \sum_{n=0}^{\infty} f(n)\rho_n^L(0\tau_0)\exp[-n(m_0+1)] \sum_{m=0}^{\infty} \binom{m+m_0}{m_0}(1-e^{-n\tau})^m = \sum_{n=0}^{\infty} f(n)\rho_n^L(0\tau_0) = \langle f(\hat{n})\rangle \tag{195}$$

The result (195a) is valid for any initial number-state Stokes beam, so we conclude that the pump beam is time-independent for arbitrary initial Stokes beam. The photon-number moments for the Stokes mode calculated from (194) are of particularly simple form. For instance, we have

$$\begin{aligned}\langle \hat{m}(\tau)\rangle &= \langle \hat{m}\rangle \sum_{n=0}^{\infty} \exp(n\,\Delta\tau)\rho_n^L(0\tau_0) + \left(\sum_{n=0}^{\infty} \exp(n\,\Delta\tau)\rho_n^L(0\tau_0) - 1\right) \\ \langle \hat{m}^2(\tau)\rangle &= (\langle \hat{m}^2\rangle + 3\langle \hat{m}\rangle + 2) \sum_{n=0}^{\infty} \exp(2n\,\Delta\tau)\rho_n^L(0\tau_0) \\ &\quad - 3(\langle \hat{m}\rangle + 1) \sum_{n=0}^{\infty} \exp(n\,\Delta\tau)\rho_n^L(0\tau_0) + 1\end{aligned} \tag{197}$$

$$\langle \hat{m}(\tau)\hat{n}(\tau)\rangle = (\langle \hat{m}\rangle + 1) \sum_{n=0}^{\infty} n \exp(n\,\Delta\tau)\rho_n^L(0\tau_0) - \langle \hat{n}\rangle \tag{198}$$

Equations (196) and (197) were obtained by Simaan.[75] Equation (196) is in agreement with the Shen relation in Ref. 26. Equations (196)–(198) reduce to Loudon's results of Ref. 30 for the simpler special case in which no scattered photons are excited initially. The sum of the mean photon numbers for the laser and Stokes mode (196) is not a constant of motion, contrary to our former considerations (132). Nonetheless, in view of the intense laser field approximation, the conservation of the total number of photons is at least approximately fulfilled. It is easy to find a physical interpretation of Eq. (196). The first term of (196) describes the amplification of the initial Stokes beam with $\langle \hat{m}\rangle$ photons at the time τ_0 and can be identified as *sensu stricto* stimulated Raman scattering. The second term of (196) corresponds to an amplification of the vacuum fluctuations and

can be interpreted as spontaneous Raman scattering, which occurs even in the case when the Stokes field contains initially no photons ($\langle \hat{m} \rangle = 0$). Note that even in the model of scattering from phonons at zero temperature ("quiet" reservoir), spontaneous scattering does take place. The coefficients $\gamma_S^{(2)}(\tau)$ and $g_{LS}^{(2)}(\tau)$, readily obtained from (35) and (38) by insertion of (196)–(198), can be explicitly compared to the coefficients calculated from other, corresponding relations. Assuming that initially there are no photons in the Stokes beam, $\langle \hat{m} \rangle = \langle \hat{m}^2 \rangle = 0$, we obtain from small-time expansions of the exponential functions in Eqs. (196) and (197) the following simple expressions for the normalized factorial moment $\gamma_S^{(2)}(\tau)$:

$$\gamma_S^{(2)}(\tau) = 2\gamma_L^{(2)} + 1 + 2\big(\langle \hat{n}^3 \rangle - \langle \hat{n}^2 \rangle^2 / \langle \hat{n} \rangle\big)\langle \hat{n} \rangle^{-2} \Delta\tau \tag{199}$$

as well as the normalized cross-correlation function $g_{LS}^{(2)}(\tau)$:

$$g_{LS}^{(2)}(\tau) = \gamma_L^{(2)} + \big(\langle \hat{n}^3 \rangle - \langle \hat{n}^2 \rangle^2 / \langle \hat{n} \rangle\big)\langle \hat{n} \rangle^{-2} \Delta\tau/2 \tag{200}$$

Equations (199) and (200) can be equivalently obtained from the short-time expansions (134) and (139), respectively, on omitting the expressions $1/\langle \hat{n} \rangle$ and $\langle \hat{n}^2 \rangle / \langle \hat{n} \rangle^2$ in the terms proportional to $\Delta\tau$, which are negligible in comparison with the terms $\langle \hat{n}^3 \rangle / \langle \hat{n} \rangle^2$ and $\langle \hat{n}^2 \rangle^2 / \langle \hat{n} \rangle^3$. The Simaan approximate relations for $g_{LS}^{(2)}(\tau)$ (32) and $\gamma_S^{(2)}(\tau)$ (33) in Ref. 75, rewritten in our notation (with extra -1 in view of (38) and (35)), do not reduce exactly to our Eqs. (199) and (200), respectively.

By substituting Eq. (194) with $\nu = \mu = 0$ into (174) one can obtain solution (176), for any initial distribution of the laser mode, with the following distribution $\rho_m^{(n_0)}(\tau)$:

$$\rho_m^{(n_0)}(\tau) = \exp[-(m+1)n_0 \Delta\tau] \times \sum_{l=0}^{m} \binom{m}{l} (e^{n_0 \Delta\tau} - 1)^l \rho_{m-l}^S(0\tau_0) \tag{201}$$

calculated for the laser field initially in a number state containing n_0 photons. In this case the mean ($\langle \hat{m}(\tau) \rangle$) and mean-square number of Stokes photons ($\langle \hat{m}^2(\tau) \rangle$),

$$\langle \hat{m}(\tau) \rangle = (\langle \hat{m} \rangle + 1)\exp(n_0 \Delta\tau) - 1 \tag{202}$$

$$\langle \hat{m}^2(\tau) \rangle = (\langle \hat{m}^2 \rangle + 3\langle \hat{m} \rangle + 2)\exp(2n_0 \Delta\tau) - 3(\langle \hat{m} \rangle + 1)\exp(n_0 \Delta\tau) + 1 \tag{203}$$

can be immediately obtained either from (196) and (197) or from (201). Assuming that the Stokes beam is initially in a coherent state $|\alpha\rangle$, we can perform summation in (201) which leads to

$$\rho_m^{(n_0)}(\tau) = \exp\left[-|\alpha|^2 - n_0\,\Delta\tau\right](1 - e^{-n_0\,\Delta\tau})^m \times {}_1F_1\left[-m; 1; -|\alpha|^2(e^{n_0\,\Delta\tau} - 1)^{-1}\right] \tag{204}$$

where ${}_1F_1$ is a confluent hypergeometric function. The density matrix elements $\rho_m^{(n_0)}(\tau_0)$ (204) describe a superposition of coherent and chaotic fields.[23, 222] This will be more transparent if we rewrite Eq. (204) in terms of the average number of Stokes photons in the chaotic part,

$$\langle \hat{m}_{\mathrm{ch}}(\tau)\rangle = \exp(n_0\,\Delta\tau) - 1 \tag{205}$$

and the mean number of photons in the coherent part alone,

$$\langle \hat{m}_c(\tau)\rangle = |\alpha|^2 \exp(n_0\,\Delta\tau) \tag{206}$$

Then, one obtains, using the Laguerre polynomial L_m, the standard form of the distribution (204)[23, 42, 75, 221]:

$$\rho_m^{(n_0)}(\tau) = \frac{\langle \hat{m}_{\mathrm{ch}}(\tau)\rangle^m}{\left(1 + \langle \hat{m}_{\mathrm{ch}}(\tau)\rangle\right)^{1+m}} \exp\left(-\frac{\langle \hat{m}_c(\tau)\rangle}{1 + \langle \hat{m}_{\mathrm{ch}}(\tau)\rangle}\right) \times L_m\left(-\frac{\langle \hat{m}_c(\tau)\rangle}{\langle \hat{m}_{\mathrm{ch}}(\tau)\rangle\left(1 + \langle \hat{m}_{\mathrm{ch}}(\tau)\rangle\right)}\right) \tag{207}$$

Similarly, by expressing the relation (202) in terms of the mean values (205) and (206) it is seen that

$$\langle \hat{m}(\tau)\rangle = \langle \hat{m}_c(\tau)\rangle + \langle \hat{m}_{\mathrm{ch}}(\tau)\rangle \tag{208}$$

The general moment of the pth order $\langle \hat{m}^p\rangle$ can be found by repeated use of the recursion relation[221]:

$$\langle m^{r+1}(\tau)\rangle = \langle m_{\mathrm{ch}}(\tau)\rangle\left(\langle m_{\mathrm{ch}}(\tau)\rangle + 1\right)\frac{\partial\langle m^r(\tau)\rangle}{\partial\langle m_{\mathrm{ch}}(\tau)\rangle} + \langle m_c(\tau)\rangle\left(2\langle m_{\mathrm{ch}}(\tau)\rangle + 1\right)\frac{\partial\langle m^r(\tau)\rangle}{\partial\langle m_c(\tau)\rangle} + \langle m^r(\tau)\rangle\langle m(\tau)\rangle \tag{209}$$

with the help of (208) or using the following explicit expression[23, 42]:

$$\langle m^r(\tau)\rangle = r!\langle m_{\rm ch}^r(\tau)\rangle L_r\left(-\frac{\langle m_c(\tau)\rangle}{\langle m_{\rm ch}(\tau)\rangle}\right) \tag{210}$$

The factorial moments of pth order can readily be calculated from (209) or (210). In particular, the second-order factorial moment reads as follows:

$$\gamma_S^{(2)}(\tau) = 1 - \left(\frac{\langle m_c(\tau)\rangle}{\langle m(\tau)\rangle}\right)^2 \tag{211}$$

which takes the minimal value, equal to zero, for the initial time τ_0, since only then $\langle \hat{m}_{\rm ch}\rangle = 0$.

For the Stokes beam initially in a vacuum state $|0\rangle$ the distributions (204) and (207) reduce to the Bose-Einstein distribution

$$\rho_m^{(n_0)}(\tau) = \frac{\langle \hat{m}_{\rm ch}(\tau)\rangle^m}{\left(1+\langle \hat{m}_{\rm ch}(\tau)\rangle\right)^{1+m}} \tag{212}$$

describing a chaotic field (cf. (14)). In this case, in the absence of stimulated scattering ($\langle \hat{m}\rangle = 0$), the chaotic field is generated in spontaneous Raman scattering as an amplification of the vacuum fluctuations.

To compare the results for the expectation values of the Stokes mode obtained in Section V.B with the present results, we assume that the laser and Stokes beams are initially in a coherent state $|\alpha_L\rangle$ and $|\alpha_S\rangle$, respectively. Performing summation in Eqs. (196)–(198) with the coherent weight function $\rho_n^L(0\tau_0)$ one readily arrives at

$$\langle \hat{m}(\tau)\rangle = \left(|\alpha_S|^2+1\right)\exp\left[|\alpha_L|^2(e^{\Delta\tau}-1)\right] - 1 \tag{213}$$

$$\langle \hat{m}^2(\tau)\rangle = \left(|\alpha_S|^4+4|\alpha_S|^2+2\right)\exp\left[|\alpha_L|^2(e^{2\Delta\tau}-1)\right] - 3\left(|\alpha_S|^2+1\right)\exp\left[|\alpha_L|^2(e^{\Delta\tau}-1)\right] + 1 \tag{214}$$

$$\langle \hat{m}(\tau)\hat{n}(\tau)\rangle = |\alpha_L|^2\left(|\alpha_S|^2+1\right)\exp\left[|\alpha_L|^2(e^{\Delta\tau}-1)+\Delta\tau\right] - |\alpha_L|^2 \tag{215}$$

Within the Fokker-Planck equation approach under parametric approximation (Section V.B) we have obtained Eqs. (111) and (114), which can be rewritten, using the notation of this section, i.e., $\kappa_S t = |e_L|^2\gamma_S t = |\alpha_L|^2\tau$,

and assuming that the mean number of phonons is zero, in the following form:

$$\langle \hat{m}(\tau)\rangle = \left(|\alpha_S|^2 + 1\right)\exp\left(|\alpha_L|^2\Delta\tau\right) - 1 \tag{216}$$

$$\langle \hat{m}^2(\tau)\rangle = \left(|\alpha_S|^4 + 4|\alpha_S|^2 + 2\right)\exp\left(2|\alpha_L|^2\Delta\tau\right) - 3\left(|\alpha_S|^2 + 1\right)\exp\left(|\alpha_L|^2\Delta\tau\right) + 1 \tag{217}$$

We also note that

$$\langle \hat{m}(\tau)\hat{n}(\tau)\rangle = \langle \hat{m}(\tau)\rangle\langle \hat{n}\rangle \tag{218}$$

For short times of evolution, $\Delta\tau \ll 1$, and intense pump beams, $|\alpha_L|^2 \gg 1$, Eqs. (213), (214), and (215) go over into Eqs. (216), 217), and (218), respectively. Indeed, the short-time expansions of (213) and (214) are

$$\langle \hat{m}(\tau)\rangle = |\alpha_S|^2 + |\alpha_L|^2\left(1 + |\alpha_S|^2\right)\Delta\tau + |\alpha_L|^2\left(|\alpha_L|^2 + 1\right)\left(1 + |\alpha_S|^2\right)\frac{(\Delta\tau)^2}{2} \tag{219}$$

$$\langle \hat{m}^2(\tau)\rangle = |\alpha_S|^2\left(1 + |\alpha_S|^2\right) + |\alpha_L|^2\left(1 + 5|\alpha_S|^2 + 2|\alpha_S|^4\right)\Delta\tau + |\alpha_L|^2\left(|\alpha_L|^2 + 1\right)\left(5 + 13|\alpha_S|^2 + 4|\alpha_S|^4\right)\frac{(\Delta\tau)^2}{2} \tag{220}$$

whereas Eqs. (216) and (217) obtained within the formalism of Section V.B reduce to

$$\langle \hat{m}(\tau)\rangle = |\alpha_S|^2 + |\alpha_L|^2\left(1 + |\alpha_S|^2\right)\Delta\tau + |\alpha_L|^4\left(1 + |\alpha_S|^2\right)\frac{(\Delta\tau)^2}{2} \tag{221}$$

$$\langle \hat{m}^2(\tau)\rangle = |\alpha_S|^2\left(1 + |\alpha_S|^2\right) + |\alpha_L|^2\left(1 + 5|\alpha_S|^2 + 2|\alpha_S|^4\right)\Delta\tau + |\alpha_L|^4\left(5 + 13|\alpha_S|^2 + 4|\alpha_S|^4\right)\frac{(\Delta\tau)^2}{2} \tag{222}$$

respectively. It is seen that for high intensity of the pump field, (219) goes over into (221), and (220) into (222) by setting $|\alpha_L|^2(|\alpha_L|^2 + 1) \approx |\alpha_L|^4$.

Hence, the factorial moment $\gamma_S^{(2)}(\tau)$,

$$\gamma_S^{(2)}(\tau) = 2|\alpha_L|^2|\alpha_S|^{-2}\Delta\tau - \left[|\alpha_L|^2\left(3 + |\alpha_S|^2\right) - |\alpha_S|^4 - 5|\alpha_S|^2 - 2\right]|\alpha_L|^2|\alpha_S|^{-4}(\Delta\tau)^2 \tag{223}$$

calculated from (219) and (220) in the case of nonzero α_S and an intense pump beam, goes over into

$$\gamma_S^{(2)}(\tau) = 2|\alpha_L|^2|\alpha_S|^{-2}\Delta\tau - \left(3 + |\alpha_S|^2\right)|\alpha_L|^4|\alpha_S|^{-4}(\Delta\tau)^2 \tag{224}$$

obtained from (221) and (222). If the initial field contains no Stokes photons, we obtain the following factorial moments $\gamma_S^{(2)}(\tau)$:

$$\gamma_S^{(2)}(\tau) = 1 + 2|\alpha_L|^{-2} + 2\,\Delta\tau + \left(2 + \tfrac{5}{6}|\alpha_L|^2\right)(\Delta\tau)^2 \tag{225}$$

$$\gamma_S^{(2)}(\tau) = 1 \tag{226}$$

within the formalisms of this section and Section V.B, respectively. The differences between the factorial moments $\gamma_S^{(2)}(\tau)$ are more pronounced in the case $\alpha_S = 0$ since the expansion of $\langle\hat{m}(\tau)\rangle$ and $\langle\hat{m}^2(\tau)\rangle$ correct to the third order in τ is required in the derivation of (225). The interbeam degree of coherence $g_{LS}^{(2)}(\tau)$ (38), as expected, vanishes for the model of Section V.B. The short-time expansion of $g_{LS}^{(2)}(\tau)$ obtained from (213)–(215), for $\alpha_S \neq 0$, is

$$g_{LS}^{(2)}(\tau) = \left(1 + |\alpha_S|^{-2}\right)\Delta\tau - \left(1 + |\alpha_S|^2\right)\left(2|\alpha_L|^2 - |\alpha_S|^2\right)|\alpha_S|^{-4}\frac{(\Delta\tau)^2}{2} \tag{227}$$

Otherwise, $\alpha_S = 0$, we get

$$g_{LS}^{(2)}(\tau) = |\alpha_L|^{-2} + \frac{1}{2}\Delta t + \frac{1}{12}\left(|\alpha_L|^2 + 3\right)\frac{(\Delta\tau)^2}{2} \tag{228}$$

It is seen that the approaches of Sections V.B and VI.B give similar predictions for the Stokes beam.

The evolution of the photon-number moments is demonstrated in Figs. 2 and 3: $\langle\hat{m}(\tau)\rangle$ calculated with Eq. (221) is depicted by solid line C or with (219) by solid line D in Fig. 2; $\langle\hat{m}^2(\tau)\rangle$ obtained from Eq. (222) is

given by solid line C or from (220) by solid line D in Fig. 3. No time dependence of $\langle \hat{n}(\tau) \rangle$ and $\langle \hat{n}^2(\tau) \rangle$ is observed for the results of this section and Section V.B; i.e., we obtain straight lines C and D in Figs. 2 and 3. Similar notation is used in Figs. 4–6 for the normalized factorial moments $\gamma_S^{(2)}(\tau)$ (Fig. 4), $\gamma_L^{(2)}(\tau)$ (Fig. 5), and the degree of interbeam coherence $g_{LS}^{(2)}(\tau)$ (Fig. 6). We have chosen rather small initial numbers of laser photons ($|\alpha_L|^2 = 2$) for numerical reasons. In this case, the factorial moments, calculated from (223), (225), (227), and (228), differ significantly from the exact numerical results. So we omit them (curves D) in Figs. 4 and 6.

VII. CONCLUSIONS

Raman scattering from a great number of phonon modes is described from a quantum-statistical point of view within the standing-wave model. The master equation for the completely quantum case, including laser pump depletion and stochastic coupling of Stokes and anti-Stokes modes, is derived and converted to classical equations: either into a generalized Fokker-Planck equation and an equation of motion for the characteristic function or into the master equation in Fock representation. These two approaches are developed both in linear and nonlinear régime. A detailed analysis of scattering into Stokes and anti-Stokes modes in linear régime, i.e., under parametric approximation, is presented. The existence of s-parametrized quasiprobability distributions, in particular the Glauber-Sudarshan P-function, is investigated. An analysis of Raman scattering into separate Stokes and anti-Stokes modes in nonlinear régime, thus including pump depletion, is given. The master equation in Fock representation is solved exactly for the complete density matrix using the Laplace transform method. Short-time solutions, steady-state solutions and approximate compact form solutions are obtained. Relations between the quasidistribution approach based on the Fokker-Planck equation and the density matrix approach based on the master equation in Fock representation are presented. The photocount distribution and its factorial moments as well as variances and extremal variances of quadratures are calculated in both approaches giving the basis for the analysis of the quantum properties of radiation such as sub-Poissonian photon-counting statistics and squeezing. A comparison of various statistical moments obtained from numerical calculations utilizing our exact solution of the master equation and from the approximate relations for short times, as well as obtained under parametric approximation, is presented graphically.

In Figs. 2–9 we compared various statistical moments obtained (1) from numerical calculations utilizing the exact solutions without the parametric approximation (Eqs. (167), (168), and/or (171) derived in Appendix B), (2) from the short-time solutions of Section VI.A.1, (3) from the solutions obtained in Section V.B within the framework of the FPE approach under the parametric approximation, and (4) from the approximate solutions derived in Section VI.B within the density-matrix formalism.

In Figs. 2, 3, 7, and 8 we demonstrated that the initial and short-time behavior of the approximate functions (curves B, C, and D) is consistent with the exact evolution (curves A). We have shown analytically that our expressions for the Stokes scattering formalisms presented in Sections V.B, VI.A.1, VI.A.2, and VI.B are equivalent for short times and high initial intensities of the pump field. Nevertheless, it is seen that the equations derived in Section VI.A.1 give the best, whereas those derived in Section VI.B give the worst approximation to the exact solution of Section VI.A.2 for small initial intensities of the laser field.

We showed in Fig. 4 (curve A) that the Stokes mode photon-number fluctuations vary from initially chaotic to Poissonian in asymptotics (for $\langle \hat{m} \rangle = 0$ and $\langle \hat{n} \rangle = |\alpha_L|^2$) or from Poissonian, through super-Poissonian, to Poissonian for large times (if $\langle \hat{m} \rangle = |\alpha_S|^2 \neq 0$ and $\langle \hat{n} \rangle = |\alpha_L|^2$). The asymptotic behavior of $\gamma_S^{(2)}(\tau)$ is consistent with our predictions in Section VI.A.3. The short-time behavior of $\gamma_S^{(2)}(\tau)$ for hyper-Raman scattering[116] is similar to that presented in Fig. 4 for Raman scattering. However, for long times the Stokes hyper-Raman photon-number fluctuations become sub-Poissonian for reasons given in Section VI.A.3.

In Fig. 5 we demonstrated that the normalized factorial moment for the laser mode, $\gamma_L^{(2)}(\tau)$, changes from initially Poissonian to super-Poissonian. The differences in $\gamma_L^{(2)}(\tau)$ between Fig. 5a (spontaneous Stokes scattering) and Fig. 5b (stimulated Stokes scattering) are only quantitative. We note that for hyper-Raman scattering the photon-number fluctuations in the initially coherent laser mode become sub-Poissonian.[116] The time behavior of the interbeam degree of coherence was presented in Figs. 6a and b. Curve A in Fig. 6a coincides with the Simaan exact solution.[75] Sub-Poissonian statistics in the compound laser-Stokes mode is observed.

It is thought (see, for instance, Ref. 22, p. 192) that the Simaan approach[75] is the most rigorous application of master equations to the Raman problem. However, Simaan's solution is restricted to the diagonal matrix elements in number representation. Only these terms are needed to obtain the mean photon numbers and their higher moments, whereby the photon correlation effects can be investigated. To investigate squeezing properties and phase correlations it is necessary to obtain the off-diagonal

elements of the density matrix. We generalized the solution of the master equation obtained by McNeil and Walls[73] and Simaan[75] to comprise all the off-diagonal matrix elements as well. Our derivation of the complete density matrix represents the main result of this paper.

Our intention was to cite an extensive literature related to our Raman scattering approaches. Nevertheless, we realize that the cited literature is not complete. We include only those references that are most relevant for the purposes of our article.

Acknowledgments

In the preparation of this paper we have benefitted greatly from contacts with Ryszard Tanaś, Przemysław Szlachetka, and Krzysztof Grygiel. Our warm appreciation must also go to Jan Peřina.

APPENDIX A

Here, we give a simple formal solution of the generalized FPE (61) for the quadidistribution $\mathscr{W}^{(-1)}(\alpha_L, \alpha_S, \alpha_A, t)$ (Q-functions) as well the corresponding characteristic function $\mathscr{C}^{(-1)}(\beta_L, \beta_S, \beta_A, t)$—the solution of the simplified equation of motion (63). We choose antinormal order ($s = -1$) to avoid the problems of existence of the quasiprobability distributions and to reduce the FPE (61) containing the third-order derivatives (for $s \neq \pm 1$) to a second-order FPE. It is by no means easy to find an exact solution of (61) or (63) even for particular orders, because the drift coefficients are not linear and the diffusion coefficients are not constant. An often employed method to solve problems of this kind is to assume that the fluctuations of the radiation fields are small compared to their mean values; i.e., the quasidistribution describing the fields is sharply peaked.[19, 23, 188, 223, 224] This will be the case for suitably chosen input state and the initial output states. Under these restrictions we can rewrite our FPE (61) related to antinormal order in the linearized form:

$$\frac{\partial}{\partial t}\mathscr{W}^{(-1)}(\alpha_L, \alpha_S, \alpha_A, t)$$

$$= \frac{1}{2}\gamma_S\left\{\left[-(D_{LS} + \xi_L\xi_S)\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_S}\right.\right.$$

$$\left. + \langle\hat{n}_S\rangle\frac{\partial}{\partial\alpha_L}\alpha_L - (\langle\hat{n}_L\rangle - 1)\frac{\partial}{\partial\alpha_S}\alpha_S + \text{c.c.}\right]$$

$$\left. + 2\langle\hat{n}_S\rangle\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_L^*}\right\}\mathscr{W}^{(-1)}$$

$$
\begin{aligned}
&+\frac{1}{2}\gamma_A\Bigg\{\Bigg[-(D_{LA}+\xi_L\xi_A)\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_A}\\
&\qquad-\left(\langle\hat{n}_A\rangle-1\right)\frac{\partial}{\partial\alpha_L}\alpha_L+\langle\hat{n}_L\rangle\frac{\partial}{\partial\alpha_A}\alpha_A+\text{c.c.}\Bigg]\\
&\qquad+2\langle\hat{n}_L\rangle\frac{\partial}{\partial\alpha_A}\frac{\partial}{\partial\alpha_A^*}\Bigg\}\mathscr{W}^{(-1)}\\
&-\Bigg\{\frac{1}{2}\gamma_{SA}e^{-2i\Delta\Omega\Delta t}\Bigg[\left(C_L+\xi_L^2\right)\left(\alpha_A^*\frac{\partial}{\partial\alpha_S}+\frac{\partial}{\partial\alpha_S}\frac{\partial}{\partial\alpha_A}-\alpha_S^*\frac{\partial}{\partial\alpha_A}\right)\\
&\qquad-2\left(\bar{D}_{SL}-\xi_L\xi_S^*\right)\frac{\partial}{\partial\alpha_L^*}\frac{\partial}{\partial\alpha_A}-\left(D_{SA}^*+\xi_S^*\xi_A^*\right)\frac{\partial^2}{\partial\alpha_L^{*2}}\Bigg]+\text{c.c.}\Bigg\}\mathscr{W}^{(-1)}\\
&+\gamma_S\langle\hat{n}_V\rangle\Bigg\{\left(\frac{1}{2}\frac{\partial}{\partial\alpha_L}\alpha_L-(D_{LS}+\xi_L\xi_S)\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_S}+\frac{1}{2}\frac{\partial}{\partial\alpha_S}\alpha_S+\text{c.c.}\right)\\
&\qquad+\langle\hat{n}_S\rangle\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_L^*}+\langle\hat{n}_L\rangle\frac{\partial}{\partial\alpha_S}\frac{\partial}{\partial\alpha_S^*}\Bigg\}\mathscr{W}^{(-1)}\\
&+\gamma_A\langle\hat{n}_V\rangle\Bigg\{\left(\frac{1}{2}\frac{\partial}{\partial\alpha_L}\alpha_L-(D_{LA}+\xi_L\xi_A)\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_A}+\frac{1}{2}\frac{\partial}{\partial\alpha_A}\alpha_A+\text{c.c.}\right)\\
&\qquad+\langle\hat{n}_A\rangle\frac{\partial}{\partial\alpha_L}\frac{\partial}{\partial\alpha_L^*}+\langle\hat{n}_L\rangle\frac{\partial}{\partial\alpha_A}\frac{\partial}{\partial\alpha_A^*}\Bigg\}\mathscr{W}^{(-1)}\\
&-\Bigg\{\gamma_{AS}\langle\hat{n}_V\rangle e^{2i\Delta\Omega\Delta t}\Bigg[\left(D_{SA}^*+\xi_S^*\xi_A^*\right)\frac{\partial^2}{\partial\alpha_L^{*2}}+\left(C_L+\xi_L^2\right)\frac{\partial}{\partial\alpha_S}\frac{\partial}{\partial\alpha_A}\\
&\qquad+\left(\bar{D}_{AL}-\xi_L\xi_A^*\right)\frac{\partial}{\partial\alpha_L^*}\frac{\partial}{\partial\alpha_S}+\left(\bar{D}_{SL}-\xi_L\xi_S^*\right)\frac{\partial}{\partial\alpha_L^*}\frac{\partial}{\partial\alpha_A}\Bigg]+\text{c.c.}\Bigg\}\mathscr{W}^{(-1)}
\end{aligned}
\tag{A.1}
$$

where the coefficients D_{kl}, $\bar{D}_{kl}$, C_k, ξ_k for $k, l = L, S, A$ are defined by (66) at the initial moment t_0. Equation (A.1) is the generalization of the FPE given in Ref. 218 for the case of nonzero γ_S and γ_{AS}. It is seen that the Raman effect under the approximations applied can be treated as an Ornstein-Uhlenbeck process.[191] The FPE (A.1) can be solved exactly by various techniques; see, e.g., Ref. 188. For instance, using the inverse Fourier transform (22), one can transform the FPE (A.1) into the corresponding equation of motion for the characteristic function

$\mathscr{E}^{(-1)}(\beta_L, \beta_S, \beta_A, t)$, which is a first-order differential equation. The method of characteristics applied to the latter equation leads to the solution

$$\begin{aligned}\mathscr{E}^{(-1)}(\beta_L, \beta_S, \beta_A, t) = \Big\langle \exp\Big\{ & - \sum_{k=L,S,A} \Big[B_k^{(-1)}(t) |\beta_k|^2 \\ & + \left(\tfrac{1}{2} C_k^*(t) \beta_k^2 + \text{c.c.}\right) + \left(\beta_k \xi_k^*(t) - \text{c.c.}\right) \Big] \\ & + \Big[D_{LS}(t) \beta_L^* \beta_S^* + \bar{D}_{LS} \beta_L \beta_S^* \\ & + D_{LA}(t) \beta_L^* \beta_A^* + \bar{D}_{LA} \beta_L \beta_A^* \\ & + D_{SA}(t) \beta_S^* \beta_A^* + \bar{D}_{SA} \beta_S \beta_A^* + \text{c.c.} \Big] \Big\} \Big\rangle \end{aligned} \tag{A.2}$$

The angle brackets mean averaging over the complex amplitudes ξ_k $(k = L, S, A)$ with the initial distribution $\mathscr{W}^{(-1)}(\alpha_L, \alpha_S, \alpha_A, t_0)$. They represent the influence of the initial photon statistics of the pump and scattered fields on the evolution of the system. The solution of Eq. (A.1) can be readily obtained by applying the Fourier transform (21) to solution (A.2), and has the form of a shifted seven-dimensional (including time) Gaussian distribution involving correlation between the radiation fields,

$$\begin{aligned}\mathscr{W}^{(-1)}(\alpha_1, \alpha_2, \alpha_3, t) = \Big\langle & \frac{1}{L^{(-1)}} \exp\Big\{ - (L^{(-1)})^{-2} \sum_{j=1}^{3} \Big[\left|\alpha_j - \xi_j(t)\right|^2 E_j^{(-1)} \\ & + \frac{1}{2} \left(\left(\alpha_j^* - \xi_j^*(t)\right)^2 E_{j+3}^{(-1)} + \text{c.c.} \right) \Big] \\ & + (L^{(-1)})^{-2} \sum_{j=1}^{2} \sum_{k=j+1}^{3} \Big[\left(\alpha_j^* - \xi_j^*(t)\right)\left(\alpha_k^* - \xi_k^*(t)\right) E_{j+k+4}^{(-1)} \\ & + \left(\alpha_j - \xi_j(t)\right)\left(\alpha_k^* - \xi_k^*(t)\right) E_{j+k+7}^{(-1)} \Big] + \text{c.c.} \Big\} \Big\rangle \end{aligned} \tag{A.3}$$

where, for simplicity, we have identified the subscripts in $\alpha_1 = \alpha_L$, $\alpha_2 = \alpha_S$, $\alpha_3 = \alpha_A$, $\xi_1 = \xi_L$, $\xi_2 = \xi_S$, $\xi_3 = \xi_A$. The functions $E_1^{(-1)}, \ldots, E_{12}^{(-1)}$, and $L^{(-1)}$, which are time-dependent, are connected with the functions

$B_k^{(-1)}, C_k, D_{kl}, \overline{D}_{kl}$ appearing in (A.2) in a manner similar to (74) and (75), respectively. We do not adduce explicit formulas for the coefficients listed, since solutions (A.2) and (A.3) serve only as an example of how one can deal with Eqs. (61) and (63). The validity of solutions (A.2) and (A.3) is restricted by strong approximations, which are actually equivalent to the parametric approximation and the short-time approximation.

APPENDIX B

Here, we solve the equation of motion (125). To eliminate the square root appearing in Eq. (125) for off-diagonal terms, it is convenient to introduce the transformation

$$\psi_{nm}(\nu\mu\tau) = \left[\frac{n!(n+\nu)!}{m!(m+\mu)!}\right]^{1/2} \rho_{nm}(\nu\mu\tau) \tag{B.1}$$

where the degree of off-diagonality μ is restricted to nonnegative integers, whereas the degree ν is $\geq -n$. On insertion of (B.1) into (125), the equation of motion for the transformed matrix elements $\psi_{nm}(\nu\mu\tau)$ takes the form

$$\begin{aligned}\psi_{nm}(\nu\mu\tau) = &-\tfrac{1}{2}[n(m+1) + (n+\nu)(m+\mu+1)]\psi_{nm}(\nu\mu\tau) \\ &+ \psi_{n+1,m-1}(\nu\mu\tau)\end{aligned} \tag{B.2}$$

We apply the Laplace transform method to (B.2), which readily leads to the solution

$$\bar{\psi}_{nm}(\nu\mu s) = \sum_{l=0}^{m} \psi_{n+l,m-l}(\nu\mu\tau_0) \prod_{p=0}^{l} [s + f(p)]^{-1} \tag{B.3}$$

for $\bar{\psi}_{nm}(\nu\mu\tau)$, the Laplace transform of $\psi_{nm}(\nu\mu\tau)$. The function $f(p)$ occurring in (B.3) is given by (169). If there are no equal terms among the elements of the set $f(0), f(1), \ldots, f(l)$ the inverse transform, after retaining the $\rho_{nm}(\nu\mu\tau)$ notation, yields (167). If there are repeated elements in the denominator of (B.3), the inverse transforms will involve convolutions. We apply two general procedures essentially equivalent to that of Simaan[75]

and Malakyan.[116] It is convenient to split (B.3) into two terms, as follows:

$$\bar{\psi}_{nm}(\nu\mu s) = \sum_{l=0}^{\lambda} \psi_{n+l,m-l}(\nu\mu\tau_0) \prod_{p=0}^{l} [s + f(p)]^{-1}$$

$$+ (1 - \delta_{m0}) \sum_{l=\lambda+1}^{m} \psi_{n+l,m-l}(\nu\mu\tau_0) \tag{B.4}$$

$$\times \prod_{p=0}^{\lambda} [s + f(p)]^{-1} \prod_{p'=\lambda+1}^{l} [s + f(p')]^{-1}$$

with λ defined by (166) (or equivalently by (170)). Let us note that a parabola $f(q) = \text{const}$ takes its maximum value for $q_0 = (2m - 2n + \mu - \nu + 2)/4$. This value, q_0 or better λ, the maximum integer $\leq q_0$, can serve as a criterion to split (B.3) in such a manner that a convolution theorem can be easily applied. The first term in (B.4) has no mutually equal factors in the denominator, so the inverse Laplace transform has the form of (167) with the proper upper limit of summation. The denominator of the second term of (B.4) contains repeated factors, which are separated, so that we can readily apply the convolution theorem finally obtaining the solution (168). Equations (167) and (168) have a rather complicated structure. We can rewrite (167) and (168) in a more compact form. If we assume that there is only one pair of equal factors among the elements of the set $f(0), f(1), \ldots, f(l)$, i.e., if

$$\bigvee_{\substack{q_1 \neq q_1' \\ q_1, q_1' \in \{0,\ldots,l\}}} f(q_1) = f(q_1') \bigwedge_{\substack{q=0,\ldots,l \\ q \neq q_1, q_1'}} f(q_1) \neq f(q)$$

then we can express the solution (B.3) for $\bar{\psi}_{nm}(\nu\mu\tau)$ as

$$\bar{\psi}_{nm}(\nu\mu s) = \sum_{l=0}^{m} \psi_{n+l,m-l}(\nu\mu\tau_0)[s + f(q_1)]^{-2} \prod_{\substack{p=0 \\ p \neq q_1, q_1'}}^{l} [s + f(p)]^{-1} \tag{B.5}$$

The inverse transform of (B.5) is

$$\psi_{nm}(\nu\mu\tau) = \sum_{l=0}^{m} \psi_{n+l,m-l}(\nu\mu\tau_0) \times \left\{ \sum_{\substack{q=0 \\ q\neq q_1, q_1'}}^{l} \exp[-f(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q}}^{l} [f(p) - f(q)]^{-1} + \left(\Delta\tau - \sum_{\substack{k=0 \\ k\neq q_1, q_1'}}^{l} [f(k) - f(q_1)]^{-1} \right) \exp[-f(q_1)\,\Delta\tau] \times \prod_{\substack{p=0 \\ p\neq q_1, q_1'}}^{l} [f(p) - f(q_1)]^{-1} \right\} \tag{B.6}$$

which is a derivative of the solution (167) over $f(q_1)$ with extra minus,

$$\psi_{nm}(\nu\mu\tau) = \sum_{l=0}^{m} \psi_{n+l,m-l}(\nu\mu\tau_0) \times \left(-\frac{\partial}{\partial f(q_1)} \right) \sum_{\substack{q=0 \\ q\neq q_1'}}^{l} \exp[-f(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q, q_1'}}^{l} [f(p) - f(q)]^{-1} \tag{B.7}$$

In the case of d equal pairs, i.e., $f(q_1) = f(q_1'), \ldots, f(q_d) = f(q_d')$, the Laplace transform solution (B.3) can be rewritten as

$$\bar{\psi}_{nm}(\nu\mu s) = \sum_{l=0}^{m} \psi_{n+l,m-1}(\nu\mu\tau_0) \times \prod_{r=1}^{d} [s + f(q_r)]^{-2} \prod_{\substack{p=0 \\ p\neq q_1, q_1', \ldots, q_d, q_d'}}^{l} [s + f(p)]^{-1} \tag{B.8}$$

finally leading to

$$\psi_{nm}(\nu\mu\tau) = \sum_{l=0}^{m} \psi_{n+l,\,m-l}(\nu\mu\tau_0)\left((-1)^d \prod_{r=1}^{d} \frac{\partial}{\partial f(q_r)}\right) \times \sum_{\substack{q=0 \\ q\neq q'_1,\ldots,q'_d}}^{l} \exp[-f(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q,q'_1,\ldots,q'_d}}^{l} [f(p) - f(q)]^{-1} \tag{B.9}$$

or equivalently to the solution (171) with the dth order differential operator $\hat{\mathscr{D}}$ (172).

APPENDIX C

In Section VI we have given an analysis of Stokes scattering. For completeness, in this appendix, we present the solution describing the anti-Stokes effect including laser depletion, but neglecting the Stokes generation and assuming that the reservoir is "quiet," i.e.,

$$\begin{aligned} \gamma_S = \gamma_{AS} = \gamma_{SA} &= 0 \\ \langle \hat{n}_V \rangle &= 0 \end{aligned} \tag{C.1}$$

Under these conditions the master equation (18) in Fock representation is

$$\frac{\partial}{\partial\tau}\rho_{nm}(\nu\mu\tau) = -\frac{1}{2}[(n+1)m + (n+\nu+1)(m+\mu)]\rho_{nm}(\nu\mu\tau) + [n(n+\nu)(m+1)(m+\mu+1)]^{1/2}\rho_{n-1,\,m+1}(\nu\mu\tau) \tag{C.2}$$

where, for brevity, we have set $n_L = n$, $n'_L = n + \nu$ (as in Section VI) and $n_A = m$, $n'_A = m + \mu$. If we define λ as follows:

$$\lambda = \left[\left[\frac{n-m+1}{2} + \frac{\nu-\mu}{4}\right]\right] \tag{C.3}$$

then the solution of (C.2) for $\lambda < 0$ becomes

$$\rho_{nm}(\nu\mu\tau) = \left[\frac{n!(n+\nu)!}{m!(m+\mu)!}\right]^{1/2} \sum_{l=0}^{n} \rho^{L}_{n-l}(\nu\tau_0)\rho^{A}_{m+l}(\mu\tau_0) \times \left[\frac{(m+l)!(m+l+\mu)!}{(n-l)!(n-l+\nu)!}\right]^{1/2} \times \sum_{q=0}^{l} \exp[-g(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q}}^{l} [g(p) - g(q)]^{-1} \tag{C.4}$$

whereas for $\lambda \geq 0$ it becomes

$$\begin{aligned}\rho_{nm}(\nu\mu\tau) = \left[\frac{n!(n+\nu)!}{m!(m+\mu)!}\right]^{1/2} \Bigg\{ & \sum_{l=0}^{\lambda} \rho^{L}_{n-l}(\nu\tau_0)\rho^{A}_{m+l}(\mu\tau_0) \times \left[\frac{(m+l)!(m+l+\mu)!}{(n-l)!(n-l+\nu)!}\right]^{1/2} \\ & \times \sum_{q=0}^{l} \exp[-g(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q}}^{l} [g(p) - g(q)]^{-1} \\ & + (1-\delta_{n0}) \sum_{l=\lambda+1}^{n} \rho^{L}_{n-l}(\nu\tau_0)\rho^{A}_{m+l}(\mu\tau_0) \times \left[\frac{(m+l)!(m+l+\mu)!}{(n-l)!(n-l+\nu)!}\right]^{1/2} \\ & \times \sum_{q=0}^{\lambda} \sum_{q'=\lambda+1}^{l} \prod_{\substack{p=0 \\ p\neq q}}^{\lambda} [g(p) - g(q)]^{-1} \prod_{\substack{p'=\lambda+1 \\ p'\neq q'}}^{l} [g(p') - g(q')]^{-1} \\ & \times \Bigg(\delta_{g(q)g(q')}\,\Delta\tau \exp[-g(q)\,\Delta\tau] \\ & + (\delta_{g(q)g(q')} - 1) \frac{\exp[-g(q)\,\Delta\tau] - \exp[-g(q')\,\Delta\tau]}{g(q) - g(q')} \Bigg) \Bigg\} \end{aligned} \tag{C.5}$$

with

$$g(x) = \tfrac{1}{2}[(m+x)(n-x+1) + (m+x+\mu)(n-x+\nu+1] \quad \text{(C.6)}$$

Alternatively, we can express solution (C.4) and (C.5) as

$$\rho_{nm}(\nu\mu\tau) = \left[\frac{n!(n+\nu)!}{m!(m+\mu)!}\right]^{1/2} \sum_{l=0}^{n} \rho^{L}_{n-l}(\nu\tau_0)\rho^{A}_{m+l}(\mu\tau_0)$$
$$\times\left[\frac{(m+l)!(m+l+\mu)!}{(n-l)!(n-l+\nu)!}\right]^{1/2} \quad \text{(C.7)}$$
$$\times\hat{\mathscr{D}} \sum_{\substack{q=0 \\ q\neq q'_1, q'_2, \ldots, q'_d}}^{l} \exp[-g(q)\,\Delta\tau] \prod_{\substack{p=0 \\ p\neq q, q'_1, q'_2, \ldots, q'_d}}^{l} [g(p) - g(q)]^{-1}$$

using the differentiation operator $\hat{\mathscr{D}}$ given by (172).

References

1. C. V. Raman and K. S. Krishnan, *Nature* **121**, 501 (1928).
2. C. V. Raman, *Indian J. Phys.* **2**, 387 (1928).
3. G. Landsberg and L. I. Mandel'stamm, *Naturwissenschaften* **16**, 557 (1928).
4. E. J. Woodbury and W. K. Ng, *Proc. IRE* **50**, 2347 (1962).
5. G. Eckhardt, R. W. Hellwarth, F. J. McClung, S. E. Schwarz, D. Weiner, and E. J. Woodbury, *Phys. Rev. Lett.* **9**, 455 (1962).
6. N. Bloembergen, *Am. J. Phys.* **35**, 989 (1967).
7. W. Kaiser and M. Maier, in F. T. Arecchi and E. O. Schultz-Dubois (Eds.), *Laser Handbook*, North-Holland, Amsterdam, 1972, p. 1077.
8. J. A. Koningstein, *Introduction to the Theory of the Raman Effect*, Reidel, Dordrecht, 1972.
9. A. Z. Grasyuk, *Kvant. Elektron.* **1**, 485 (1974) [*Sov. J. Quantum Electron.* **4**, 269 (1974)].
10. A. Z. Grasyuk, in N. G. Basov (Ed.), Lasers and Their Applications, Proc. of the Lebedev Physics Institute, Vol. 76, Nauka, Moscow, 1976.
11. C. S. Wang, in M. Rabin and C. L. Tang (Eds.), *Quantum Electronics: A Treatise*, Vol. 1, Part A, Academic, New York, 1975, p. 447.
12. C. S. Wang, *Phys. Rev.* **182**, 482 (1969).
13. M. Cardona, (Ed.), *Light Scattering in Solids, Topics Appl. Phys.*, Vol. 8, Springer, Berlin, 1975.
14. D. A. Long, *Raman Spectroscopy*, McGraw-Hill, New York, 1977.
15. W. Hayes and R. Loudon, *Scattering of Light by Crystals*, Wiley, New York, 1978.
16. A. Penzkofer, A. Laubereau, and W. Kaiser, *Prog. Quantum Electron.* **6**, 55 (1979).
17. S. Kielich, *Prog. Opt.* **20**, 156 (1983).
18. S. Kielich, *Proc. Indian Acad. Sci. (Chem. Sci.)* **94**, 403 (1985).

19. S. Kielich, *Nonlinear Molecular Optics*, Nauka, Moscow, 1981 (in Russian).
20. Y. R. Shen, *Nonlinear Optics*, Wiley, New York, 1984, Chapter 10.
21. Yu. E. D'yakov and S.Yu. Nikitin, *Kvant. Elektron.* **14**, 1925 (1987) [*Sov. J. Quantum Electron.* **17**, 1227 (1987)].
22. M. G. Raymer and I. A. Walmsley, *Prog. Opt.* **28**, 182 (1990).
23. J. Peřina, *Quantum Statistics of Linear and Nonlinear Optical Phenomena*, Reidel, Dortrecht, 1984 (2d enlarged ed., 1991).
24. J. Mostowski and M. G. Raymer, in G. P. Agrawal and W. Boyd (Eds.), in *Contemporary Nonlinear Optics*, Academic, Boston, 1992, p. 187.
25. Special issue of *J. Opt. Am. Soc. B* **3** (10) (1986).
26. Y. R. Shen, *Phys. Rev.* **155**, 921 (1967).
27. P. Szlachetka, *Quantum Fluctuations in Multi-Photon Raman Scattering Processes*, Thesis, Poznań University, 1980.
28. I. Abram, *Phys. Rev. A* **35**, 4661 (1987).
29. I. Abram and E. Cohen, *Phys. Rev. A* **44**, 500 (1987).
30. R. Loudon, *The Quantum Theory of Light*, Clarendon, Oxford, UK, 1973 (2nd enlarged ed., 1983).
31. A. Schenzle and H. Brand, *Phys. Rev. A* **20**, 1628 (1979).
32. C. Mavroyannis, *Mol. Phys.* **48**, 4847 (1983).
33. C. Mavroyannis, *J. Phys. Chem.* **88**, 4868 (1984).
34. C. Mavroyannis, *J. Chem. Phys.* **47**, 2554 (1986).
35. H. S. Freedhoff, *J. Chem. Phys.* **47**, 2554 (1967).
36. N. Bloembergen and Y. R. Shen, *Phys. Rev. Lett.* **13**, 720 (1964).
37. N. Bloembergen and Y. R. Shen, *Phys. Rev.* **133**, A37 (1964).
38. Y. R. Shen and N. Bloembergen, *Phys. Rev.* **137**, A1787 (1965).
39. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan *Phys. Rev.* **127**, 1918 (1962).
40. E. A. Mishkin and D. F. Walls, *Phys. Rev.* **185**, 1618 (1969).
41. W. H. Louisell, A. Yariv, and A. E. Siegman, *Phys. Rev.* **124**, 1646 (1961).
42. B. R. Mollow and R. J. Glauber, *Phys. Rev. A* **160**, 1076, 1097 (1967).
43. R. Graham and H. Haken, *Z. Phys.* **210**, 276 (1968).
44. J. Tucker and D. F. Walls, *Phys. Rev.* **178**, 2036 (1969).
45. M. E. Smithers and E. Y. C. Lu, *Phys. Rev. A* **10**, 1874 (1974).
46. S. K. Srinivasan and S. Udayabaskaran, *Opt. Acta* **26**, 1535 (1979).
47. V. Peřinová, *Opt. Acta* **28**, 747 (1981).
48. V. Peřinová and J. Peřina, *Opt. Acta* **28**, 769 (1981).
49. C. M. Caves and B. L. Schumaker, *Phys. Rev. A* **31**, 3068 (1985).
50. B. L. Schumaker and C. M. Caves, *Phys. Rev. A* **31**, 3093 (1985).
51. D. F. Walls, *Z. Phys.* **234**, 231 (1970).
52. D. F. Walls, *J. Phys. A* **4**, 813 (1971).
53. J. Peřina, *Opt. Acta* **28**, 325 (1981).
54. J. Peřina, *Opt. Acta* **28**, 1529 (1981).
55. J. Peřina, *Acta Phys. Pol. A* **78**, 173 (1990).

56. M. Kárská and J. Peřina, *J. Mod. Opt.* **37**, 195 (1990).
57. D. F. Walls, *Z. Phys.* **237**, 224 (1970).
58. P. Szlachetka, S. Kielich, J. Peřina, and V. Peřinová, *J. Phys. A* **12**, 1921 (1979).
59. P. Szlachetka, S. Kielich, J. Peřina, and V. Peřinová, *J. Mol. Spectrosc.* **61**, 281 (1980).
60. P. Szlachetka, S. Kielich, J. Peřina, and V. Peřinová, *Opt. Acta* **27**, 1609 (1980).
61. P. Szlachetka and S. Kielich, in W. F. Murphy (Ed.), *Photon Co- and Anti-correlations in Two-photon Raman Scattering*, VII International Conference on Raman Spectroscopy, Ottawa, 1980, North Holland, Amsterdam, 1980.
62. T. V. Trung and F.-J. Schütte, *Ann. Phys. (Leipzig)* **7**, 216 (1978).
63. W. Tänzler and F.-J. Schütte, *Ann. Phys. (Leipzig)* **7**, 73 (1981).
64. F. G. Reis and M. P. Sharma, *Opt. Commun.* **41**, 341 (1982).
65. J. Peřina, V. Peřinová, and J. Kodousek, *Opt. Commun.* **49**, 210 (1984).
66. J. Peřinová, M. Kárská, and J. Křepelka, *Acta Phys. Pol. A* **79**, 817 (1991).
67. J. Peřina and J. Křepelka, *J. Mod. Opt.* **38**, 2137 (1991).
68. J. Peřina and J. Křepelka, *J. Mod. Opt.* **39**, 1029 (1992).
69. M. D. Levenson, M. J. Holland, D. F. Walls, P. J. Manson, P. T. H. Fisk, and H. A. Bachor, *Phys. Rev. A* **44**, 2023 (1991).
70. G. P. Agrawal and C. L. Mehta, *J. Phys. A* **7**, 607 (1974).
71. M. Schubert and B. Wilhelmi, *Nonlinear Optics and Quantum Electronics*, Wiley, New York, 1986, Chapter 1.
72. Y. R. Shen, in R. J. Glauber (Ed.), *Quantum Theory of Nonlinear Optics*, Proc. Int. School Phys. "Enrico Fermi," Vol. 42, Academic, New York, 1969, p. 473.
73. K. J. McNeil and D. F. Walls, *J. Phys. A* **7**, 617 (1974).
74. D. F. Walls, *Z. Phys.* **244**, 117 (1971).
75. H. D. Simaan, *J. Phys. A* **8**, 1620 (1975).
76. H. Iwasawa, *Z. Phys.* **23**, 399 (1976).
77. D. Eimerl, *Phys. Rev. A* **20**, 369 (1979).
78. P. S. Gupta and B. K. Mohanty, *Czech. J. Phys. B* **30**, 1127 (1980).
79. B. K. Mohanty and P. S. Gupta, *Czech. J. Phys. B* **31**, 275 (1981).
80. B. K. Mohanty, N. Nayak, and P. S. Gupta, *Opt. Acta* **29**, 1017 (1982).
81. N. N. Bogolubov, Jr., F. Le Kien, and A. S. Shumovsky, *Physica* **130A**, 273 (1985).
82. K. Grygiel and S. Kielich, *J. Mod. Opt.* **34**, 61 (1987).
83. K. Grygiel, *Quantum Theory of Stimulated Raman Scattering Processes* Thesis, Poznań University, 1987 (in Polish).
84. V. A. Andreev and P. B. Lerner, *Opt. Commun.* **84**, 323 (1991).
85. Th. Hansch and P. Toschek, *Z. Phys.* **236**, 213 (1970).
86. P. G. Brewer and E. L. Hahn, *Phys. Rev. A* **11**, 1641 (1975).
87. G. S. Agarwal and S. Jha, *Z. Phys. B* **35**, 391 (1979).
88. N. N. Bogolubov, Jr., A. S. Shumovsky, and T. Quang, *Physica* **144A**, 503 (1987).
89. A. S. Shumovsky, T. Quang, and R. Tanaś, *Physica* **149A**, 283 (1988).
90. P. S. Gupta and J. Dash, *Czech. J. Phys.* **40**, 432 (1990).
91. A. A. Villaeys, A. Boeglin, and S. H. Lin, *Phys. Rev. A* **44**, 4660, 4671 (1991).
92. H. Ritsch, M. A. M. Marte, and P. Zoller, *Europhys. Lett.* **19**, 7 (1992).

93. S. Kielich and P. Piątek, The squeezed states of fields in propagation and harmonic generation of laser beam, paper of this compilation, Part I, 1993.
94. S. Kielich, R. Tanaś, and R. Zawodny, Self-squeezing of elliptically polarized light propagating in a Kerr-like optically active medium, paper of this compilation, Part I, 1993.
95. J. Peřina, J. Bajer, V. Peřinová, and Z. Hradil, Photon statistics of nonclassical fields, paper of this compilation, Part I, 1993.
96. S. Kielich, M. Kozierowski, and R. Tanaś, *Opt. Acta* **32**, 1023 (1985).
97. G. Leuchs, in G. T. Moore and M. O. Scully (Eds.), *Frontiers of Nonequilibrium Statistical Physics*, Plenum, New York, 1986.
98. R. Loudon and P. L. Knight, *J. Mod. Opt.* **34**, 709 (1987).
99. M. C. Teich and B. E. A. Saleh, *Prog. Opt.* **26**, 1 (1988).
100. M. C. Teich and B. E. A. Saleh, *Quantum Opt.* **1**, 153 (1989).
101. K. Zaheer and M. S. Zubairy, *Adv. At. Mol. Opt. Phys.* **28**, 143 (1990).
102. Special issue of *J. Opt. Soc. Am. B* **4** (10) (1987).
103. Special issue of *J. Mod. Opt.* **34** (6/7) (1987).
104. K. Germey, F.-J. Schütte, and R. Tiebel, *Ann. Phys. (Leipzig)* **38**, 80 (1981).
105. P. Král, *Czech. J. Phys.* **40**, 1226 (1990).
106. P. S. Gupta and J. Dash, *Indian J. Pure and Appl. Phys.* **29**, 606 (1991).
107. H. D. Simaan, *J. Phys. A* **11**, 799 (1978).
108. P. Szlachetka and S. Kielich, *VIII Conf. on Quantum Electronics and Nonlinear Optics*, Poznań, 24–27 April 1978, Section B, p. 33.
109. G. S. Agarwal, *Opt. Commun.* **31**, 325 (1979).
110. V. Peřinová, J. Peřina, P. Szlachetka, and S. Kielich, *Acta Phys. Pol. A* **56**, 267, 275 (1979).
111. W. Tänzler and F.-J. Schütte, *Opt. Commun.* **37**, 447 (1981).
112. B. K. Mohanty and P. S. Gupta, *Czech. J. Phys. B* **31**, 857 (1981).
113. V. Peřinová and R. Tiebel, *Opt. Commun.* **50** 401 (1984).
114. P. S. Gupta and J. Dash, *Opt. Commun.* **88**, 273 (1992).
115. Yu.P. Malakyan, *Opt. Commun.* **78**, 67 (1990).
116. Yu.P. Malakyan, *J. Mod. Opt.* **39**, 509 (1992).
117. S. Kielich, *J. Phys. Lett.* **43**, L-389 (1982).
118. S. Singh, *Opt. Commun.* **44**, 254 (1983).
119. X. T. Zhou and L. Mandel, *Phys. Rev. A* **41**, 475 (1990).
120. M. Le Berre-Rousseau, E. Ressayre, and A. Tallet, *Phys. Rev. Lett.* **43**, 1314 (1979).
121. Z. Bialynicka-Birula, I. Bialynicki-Birula, and G. M. Salomone, *Phys. Rev. A* **43**, 3696 (1991).
122. D. F. Walls, *J. Phys. A* **6**, 496 (1973).
123. M. Smoluchowski, *Ann. Phys.* **25**, 332 (1908).
124. P. D. Drummond and C. W. Gardiner, *J. Phys. A* **13**, 2353 (1980).
125. S. J. Carter and P. D. Drummond, *Phys. Rev. Lett.* **67**, 3757 (1991).
126. N. R. Davis and C. D. Cantrell, *J. Opt. Am. Soc. B* **6**, 74 (1989).
127. S. L. Braunstein, C. M. Caves, and G. J. Milburn, *Phys. Rev. A* **43**, 1153 (1991).

128. R. Schack and A. Schenzle, *Phys. Rev. A* **44**, 682 (1991).
129. D. T. Pegg and S. M. Barnett, *Phys. Rev. A* **39**, 1665 (1989).
130. R. Tanaś, Ts. Gantsog, A. Miranowicz, and S. Kielich, *J. Opt. Soc. Am. B* **8**, 1576 (1991).
131. Ts. Gantsog, A. Miranowicz, and R. Tanaś, *Phys. Rev A* **46**, 2870 (1992).
132. R. Tanaś, A. Miranowicz, and S. Kielich, *Phys. Rev. A* **43**, 4014 (1991).
133. P. L. Knight, *Phys. Scr. T* **12**, 51 (1986).
134. S. J. D. Phoenix and P. L. Knight, *J. Opt. Soc. Am. B* **7**, 116 (1990).
135. L. Schoendorff and H. Risken, *Phys. Rev. A* **41**, 5147 (1990).
136. G. S. Agarwal and R. R. Puri, *Phys. Rev. A* **43**, 3949 (1991).
137. R. R. Puri and G. S. Agarwal, *Phys. Rev. A* **45**, 5073 (1992).
138. C. C. Gerry and J. H. Eberly, *Phys. Rev. A* **42**, 6805 (1990).
139. C. C. Gerry, *Phys. Lett. A* **161**, 9 (1991).
140. C. C. Gerry and H. Huang, *Phys. Rev. A* **45**, 8037 (1992).
141. G. Gangopadhyay and D. S. Ray, *Phys. Rev. A* **45**, 1843 (1992).
142. J. Mostowski and M. G. Raymer, *Opt. Commun.* **36**, 237 (1981).
143. M. G. Raymer and J. Mostowski, *Phys. Rev. A* **24**, 1980 (1981).
144. S. Ya. Kilin, *Europhys. Lett.* **5**, 419 (1988).
145. Z. Z. Li, C. Radzewicz, and M. G. Raymer, *J. Opt. Soc. Am. B* **5**, 2340 (1988).
146. S. A. Akhmanov, A. P. Sukhorukov, and A. S. Chirkin, *Zh. Eksp. Teor. Fiz.* **55**, 1430 (1968) [*Sov. Phys.-JETP* **28**, 748 (1969)].
147. T. von Foerster and R. J. Glauber, *Phys. Rev. A* **3**, 1484 (1971).
148. S. A. Akhmanov, K. N. Drabovich, A. P. Sukhorukov and A. S. Chirkin, *Zh. Eksp. Teor. Fiz.* **59**, 485 (1970) [*Sov. Phys.-JETP* **32**, 266 (1971)].
149. V. I. Emel'yanov and V. N. Seminogov, *Zh. Eksp. Teor. Fiz.* **76**, 34 (1979) [*Sov. Phys.-JETP* **49**, 17 (1979)].
150. E. Haake, H. King, G. Schröder, J. W. Haus, and R. Glauber, *Phys. Rev. A* **20**, 2047 (1979); **23**, 1322 (1981).
151. M. G. Raymer, K. Rzążewski, and J. Mostowski, *Opt. Lett.* **7**, 71 (1982).
152. F. Haake, *Phys. Lett. A* **90**, 127 (1982).
153. J. Mostowski and B. Sobolewska, *Phys. Rev. A* **30**, 610 (1984).
154. I. A. Walmsley and M. G. Raymer, *Phys. Rev. Lett.* **50**, 962 (1983).
155. M. G. Raymer and I. A. Walmsley, in L. Mandel and E. Wolf (Eds.), *Proc. 5th Rochester Conf. on Coherence and Quantum Optics*, Plenum, New York, 1984.
156. N. Fabricius, K. Nattermann, and D. van der Linde, *Phys. Rev. Lett.* **52**, 113 (1984).
157. D. C. MacPherson, R. C. Swanson, and J. L. Carlsten, *Phys. Rev. A* **40**, 6745 (1989).
158. R. C. Swanson, D. C. MacPherson, P. R. Battle, and J. L. Carlsten, *Phys. Rev. A* **45**, 450 (1992).
159. J. C. Englund and C. M. Bowden, *Phys. Rev. Lett.* **57**, 2661 (1986).
160. J. C. Englund and C. M. Bowden, *Phys. Rev. A* **42**, 2870 (1990).
161. S. G. Rautian and B. M. Chernobrod, *Zh. Eksp. Teor. Fiz.* **72**, 1342 (1977) [*Sov. Phys.-JETP* **54**, 642 (1977)].

162. Yu. M. Kirin, Yu. N. Popov. S. G. Rautian, V. P. Safonov, and B. M. Chernobrod, *Kvant. Elektron.* **1**, 430 (1974) [*Sov. J. Quantum Electron.* **4**, 244 (1974)].
163. V. S. Pivtsov, S. G. Rautian, V. P. Safonov, K. G. Folin, and B. M. Chernobrod, *JETP Lett.* **30**, 317 (1979); Zh. Eksp. Teor. Fiz. **81**, 468 (1981) [Sov. Phys.-JETP **54**, 250 (1981)].
164. R. W. Terhune, P. D. Maker, and C. M. Savage, *Phys. Rev. Lett.* **14**, 681 (1965).
165. P. D. Maker, *Physics and Quantum Electronics*, McGraw-Hill, New York, 1966, p. 60.
166. T. Neugebauer, *Acta. Phys. Hungar.* **16**, 217, 227 (1963).
167. S. Kielich, *Bull. Acad. Pol. Sci.* (*Sér. Math. Astron. Phys.*) **11**, 201 (1963); **12**, 53 (1964).
168. S. Kielich, *Acta Phys. Pol.* **26**, 135 (1964).
169. Y. Li, *Acta Phys. Sinica* **20**, 164 (1964).
170. S. Kielich, *Proc. Phys. Soc. London* **86**, 709 (1965).
171. S. Kielich, *Acta Phys. Pol.* **30**, 393 (1966).
172. D. C. Hanna, M. A. Yuratich, and D. Cotter, *Nonlinear Optics of Free Atoms and Molecules*, Springer, Heidelberg, 1979.
173. Ch. Kittel, *Quantum Theory of Solids*, Wiley, New York, 1963.
174. R. H. Pantell and H. E. Puthoff, *Fundamentals of Quantum Electronics*, Wiley, New York, 1969.
175. H. H. Ritze and A. Bandilla, *Opt. Commun.* **29**, 126 (1979).
176. R. Tanaś and S. Kielich, *Opt. Commun.* **30**, 443 (1979).
177. R. Tanaś and S. Kielich, *Opt. Commun.* **45**, 351 (1983).
178. R. Tanaś and S. Kielich, *Opt. Acta* **31**, 81 (1984).
179. Ts. Gantsog and R. Tanaś, *J. Mod. Opt.* **38**, 1537 (1991).
180. R. J. Glauber, *Phys. Rev.* **130**, 2529 (1963).
181. G. Rivoire, *C. R. Acad. Sci. Paris* **260**, 5743 (1965).
182. J. Peřina, J. Peřinová, P. Szlachetka, and S. Kielich, *Approximate Photon and Phonon Statistics and Anticorrelation in Raman and Brillouin Scattering*, Commun. Palacký University, Olomouc, 1979.
183. W. H. Louisell, *Quantum Statistical Properties of Radiation*, Wiley, New York, 1973.
184. C. W. Gardiner, in H. Haken (Ed.), *Springer Series in Synergetics*, Vol. 13, *Handbook of Stochastic Methods*, Springer, Berlin, 1984.
185. G. S. Agarwal, *Prog. Opt.* **11**, 1 (1973).
186. H. Haken, *Handbuch der Physik*, Vol. 25/2c, Springer, Berlin, 1970.
187. M. W. Evans, G. J. Evans, W. T. Coffey, and P. Grigolini, *Molecular Dynamics and Theory of Broad Band Spectroscopy*, Wiley, New York, 1982.
188. H. Risken, *The Fokker-Planck Equation*, Springer, Berlin, 1984.
189. V. Peřinová, J. Křepelka, and J. Peřina, *Opt. Acta* **33**, 1263 (1986).
190. M. Sugawara, M. Hayashi, and Y. Fujimura, *Chem. Phys. Lett.* **189**, 346 (1992).
191. G. E. Uhlenbeck, L. S. Ornstein, *Phys. Rev.* **36**, 823 (1930).
192. R. J. Glauber, *Phys. Rev. A* **131**, 2766 (1963).
193. R. J. Glauber, *Phys. Rev. Lett.* **10**, 84 (1963).
194. K. E. Cahill, *Phys. Rev.* **138**, B1566 (1965).
195. J. R. Klauder, J. McKenna, and D. G. Currie, *J. Math. Phys.* **6**, 743 (1965).

196. G. S. Agarwal and E. Wolf, *Phys. Lett.* **26A**, 485 (1968).
197. K. E. Cahill and R. J. Glauber, *Phys. Rev.* **177**, 1857, 1882 (1969).
198. J. R. Klauder and E. C. G. Sudarshan, *Fundamentals of Quantum Optics*, Benjamin, New York, 1968.
199. V. I. Tatarsky, *Usp. Fiz. Nauk* **139**, 587 (1983).
200. M. Hillery, R. F. O'Connell, M. O. Scully, and E. P. Wigner, *Phys. Rep.* **106**, 121 (1984).
201. A. Miranowicz, R. Tanaś and S. Kielich, *Quantum Opt.* **2**, 253 (1990).
202. U. M. Titulaer and R. J. Glauber, *Phys. Rev.* **140**, B676 (1965).
203. M. Hillery, *Phys. Rev. A* **35**, 725 (1987).
204. S. Stenholm, *Eur. J. Phys.* **1**, 244 (1980).
205. L. Mandel, *Proc. Phys. Soc.* **72**, (1958) 1037; **74**, 233 (1959).
206. L. Mandel, *Prog. Opt.* **2**, 181 (1963).
207. J. Peřina and R. Horak, *Opt. Commun.* **1**, 91 (1969).
208. R. J. Glauber, in C. DeWitt, A. Blandin, and C. Cohen-Tannoudji (Eds.), *Quantum Optics and Electronics*, Gordon & Breach, New York, 1965, p. 144.
209. B. Saleh, *Springer Ser. Opt. Sci., Vol.* 6 *Photoelectron Statistics*, Springer, Berlin, 1978.
210. H. Paul, *Rev. Mod. Phys.* **54**, 1061 (1982).
211. L. Lukš, V. Peřinová, and J. Peřina, *Opt. Commun.* **67**, 149 (1988).
212. R. Loudon, *Opt. Commun.* **70**, 109 (1989).
213. A. Lukš, V. Peřinová, and Z. Hradil, *Acta Phys. Pol. A* **74**, 713 (1988).
214. E. Schrödinger, *Sitzungsber. Preuss. Akad. Wiss. Phys. Math. K* **1**, 296 (1930).
215. M. Kozierowski and V. I. Man'ko, *Opt. Commun.* **69**, 71 (1988).
216. K. Vogel and H. Risken, *Phys. Rev. A* **39**, 4675 (1989).
217. Ning-Lu, Shi-Yao Zhu, and G. S. Agarwal, *Phys. Rev. A* **40**, 258 (1989).
218. A. Miranowicz, *Master Equation Approach to Raman Scattering*, Thesis, Poznań University, 1988 (in Polish).
219. H. Fearn and M. J. Collet, *J. Mod. Opt.* **35**, 553 (1988).
220. M. O. Scully and W. E. Lamb, *Phys. Rev.* **159**, 208 (1967).
221. G. Lachs, *Phys. Rev.* **138**, B1012 (1965).
222. V. Peřinová, A. Lukš, and P. Szlachetka, *J. Mod. Opt.* **36**, 1435 (1989).
223. D. A. Long and L. Stanton, *Proc. R. Soc. London Ser. A* **318**, 441 (1970).
224. S. M. Barnett, S. Stenholm, and D. T. Pegg, *Opt. Commun.* **73**, 314 (1989).