PHYSICAL REVIEW A VOLUME 30, NUMBER 5 NOVEMBER 1984

Stochastic pump effects in lasers

R. F. Fox, G. E. James, and R. Roy
School of Physics, Georgia Institute of Technology, Atlanta, Georgia 30332
(Received 18 May 1984)

We examine from first principles the effects of a stochastic pump on the output fluctuations of a laser. The Hamiltonian for a four-level molecule interacting with a stochastic semiclassical pump field and quantized laser field is used to obtain density-matrix equations of motion. Noise from the pump field and from spontaneous emission is introduced systematically, as opposed to the *ad hoc* inclusion of random forces in the semiclassical equations. The relationship of this theory with previous quantum and semiclassical equations is described. New equations are derived on which computer simulations are performed to compare our theoretical results to experimental data for a single-mode dye laser.

I. INTRODUCTION

During the last few years it has become apparent that the presence of stochasticity in the pump source may drastically alter the coherence properties of a laser. Apart from an early study of this problem by Wang and Lamb,[1] no systematic study of this problem appears to have been carried out. Recent research on the subject has been based on the *ad hoc* inclusion of noise terms in the semiclassical Lamb equation.[2–4] A new, comprehensive theory, based on first principles, is clearly required if one is to understand the transformation of the pump noise by the nonlinear laser system and include quantum effects such as spontaneous emission. It is the purpose of this paper to present such a theory, starting from the Hamiltonian for a four-level molecule interacting simultaneously with a stochastic semiclassical pump field and quantized laser field.

As long as ten years ago, there appeared to be ample evidence that conventional laser theory[5] and experiments[6] on the coherence properties on lasers were in good agreement with each other. Experiments on single mode dye lasers by Mandel and his co-workers[7] demonstrated, however, that fluctuation phenomena in these lasers showed very different characteristics than those expected on the basis of standard theory.

Kaminishi *et al.* attempted to apply the Haken-Risken theory[5(a),5(b)] for single mode lasers near threshold to their measurements on single mode dye lasers operated near threshold. This theory is basically a Langevin approach to laser noise governed by the equation

$$\dot{E}(t)=(a-A\,|E\,|^2)E+\xi(t) \tag{1}$$

in which $E(t)$ is the complex amplitude of the laser electric field, a and A are real parameters. a is the pump parameter whereas $A(>0)$ provides stabilization above threshold as a result of saturation of the laser active medium. $\xi(t)$ is a Gaussian, white-noise source, representing the contribution of spontaneous emission, and is of *ad hoc* origin.

Kaminishi *et al.* found that this theory was inadequate in explaining the behavior of the observed relative mean-square intensity fluctuations (versus pump parameter) or the measured form of the intensity correlation functions. Realizing that pump laser fluctuations could play a role of importance, they suggested a preliminary treatment of the mean-square intensity fluctuations which included this source of stochasticity. Another possibility was the effect of triplet state absorption which may occur in organic dyes. This situation was considered from a quantum-mechanical viewpoint in the paper of Schaefer and Willis.[8] They gave an elegant treatment of the effect of the triplet states on dye laser fluctuations. The conditions of operation of the dye laser on which measurements were made indicated, though, that these triplet state effects were not dominant since a triplet quenching agent was used in the dye solution.

In order to explain the measurements, Graham *et al.*[2] made the simple assumption that the pump parameter a is noisy. They then dropped $\xi(t)$ from Eq. (1) and replaced a with a Gaussian, white-noise process. This converts Eq. (1) from an additive stochastic process to a multiplicative process,[9] one which can be solved exactly.[10] With this approach, they were able to fit the published data of Kaminishi *et al.*[7] Unfortunately, this analysis did not fit some unpublished data, as was pointed out by Short *et al.*[3] in a subsequent paper. They suggested the problem with fitting the data may lie in the need to use "colored" noise rather than white noise. This means that the time scale for the relaxation of the pump noise may not be short enough compared with all other time scales characterizing the dye laser so that it is not a good approximation to use a Dirac δ-function correlation for the pump noise autocorrelation function.

The drawback to using colored noise, however, is that the equations are no longer analytically solvable. The formal theory for colored noise approximations is well developed[11] but only rarely leads to exact analytic solutions. The equation studied in Refs. (2) and (10),

$$\dot{E}(t)=[a(t)-A\,|E\,|^2]E \tag{2}$$

for which the authors found exact solutions, is no longer tractable if $a(t)$ contains colored noise. Dixit and Sahni,[4] assuming an exponentially correlated noise source, performed computer simulations to obtain fits to the measured intensity correlations of Short *et al.*[3] allowing the correlation time for the pump noise to be an arbitrary parameter. They also had to adjust the value of the relative mean-squared fluctuations to obtain reasonable fits. This reflects the *ad hoc* nature of the inclusion of noise in Eq. (2).

It is tempting to consider the subject closed at this point. However, if one examines Eq. (2), one notices several inadequacies in the justification of its basic form.

First and foremost, Eq. (2) is phenomenological. If it is correct, one should be able to derive it on more basic grounds. Second, quantum noise has been totally neglected in this equation. Third, if stochastic pump effects are important, not only the pump parameter a but also the saturation parameter A should contain random fluctuations.[1] These inadequacies represent serious defects in the theories reviewed so far. We address each of these problems here.

The remainder of the paper is organized as follows. In Sec. II, the description of a laser at the level of the quantum-mechanical density matrix for the active molecule energy levels and the photon quanta for the laser field is given. The stochastic pump field is treated semiclassically. Decay transitions between molecular energy levels and cavity relaxation are modeled by stochastic interactions, following a method developed by Fox.[9] In Sec. II, adiabatic elimination is used to remove off-diagonal density matrix elements. A photon state contraction is performed in Sec. IV. The correspondence of our theory with semiclassical equations is presented in Sec. V, along with the results of computer simulations. In Sec. VI, an analytic approximation is examined which includes quantum spontaneous emission effects. The analysis of the equations containing both quantum fluctuations and stochastic pump effects is somewhat involved, and will be discussed in a future publication. In Sec. VII we summarize our results and assess the status of the entire problem of stochastic pump effects in lasers.

II. DENSITY-MATRIX DYNAMICS

The laser active medium will be modeled as a four-level molecular system in a resonant cavity. Figure 1 shows a

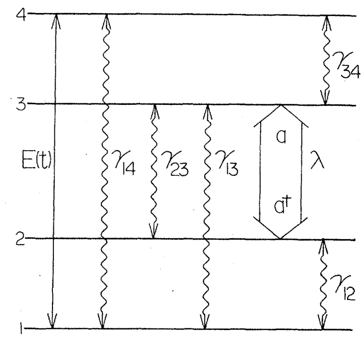

FIG. 1. Dye molecule energy levels interacting with pump and laser radiation fields. The upward pointing arrows for the stochastic transitions are in fact suppressed by Boltzmann factors.

schematic diagram of the molecular energy levels as well as the various transition mechanisms. The ground state is level 1. The pump transition is from level 1 to level 4, which is resonant with the pump laser. The wiggles show transitions due to collisions or radiative transitions not related to the pump or lasing radiation. The decay process connecting levels 4 and 3 and levels 2 and 1 are very fast. The thick arrow between levels 3 and 2 is the lasing transition which yields the light for which the laser cavity is tuned. We will consider only single-mode operation in this paper. The pump field will be treated semiclassically, whereas the laser field will be quantized in the following treatment.

The Hilbert space relevant for the density-matrix description of the four-level molecule and the quantized laser field is a direct product of the four-dimensional Hilbert space of the molecular levels and the infinite dimensional Hilbert space of the quantized field. The density-matrix equation is (in which $[H,\cdot]=H\cdot-\cdot H$ is a "commutator" operator)

$$i\hbar\frac{\partial\rho}{\partial t}=[H,\rho] \tag{3}$$

in which ρ is the density matrix for the molecular levels and the photons and H is the total Hamiltonian

$$\begin{aligned}H=&\hbar\omega_{23}a^\dagger a+\sum_{i=1}^{4}|i\rangle\epsilon_i\langle i|+\mu\cos(\omega_{14}t)(|4\rangle E_p(t)\langle 1|+|1\rangle E_p^*(t)\langle 4|)+g(a+a^\dagger)(|3\rangle\langle 2|+|2\rangle\langle 3|)\\&+\widetilde{H}_{34}(t)(|3\rangle\langle 4|+|4\rangle\langle 3|)+\widetilde{H}_{14}(t)(|1\rangle\langle 4|+|4\rangle\langle 1|)\\&+\widetilde{H}_{23}(t)(|2\rangle\langle 3|+|3\rangle\langle 2|)+\widetilde{H}_{13}(t)(|1\rangle\langle 3|+|3\rangle\langle 1|)+\widetilde{H}_{12}(t)(|1\rangle\langle 2|+|2\rangle\langle 1|)+\widetilde{H}_c(t)\ .\end{aligned} \tag{4}$$

Strictly, those parts of this Hamiltonian which act in only one factor of the product Hilbert space should be written as a direct product with an identity operator for the other Hilbert space factor, e.g.,

$$\hbar\omega_{23}a^\dagger a\equiv\hbar\omega_{23}aa^\dagger\otimes 1_M\ . \tag{5}$$

We will dispense with such cumbersome notation. The first term in (4) is the Hamiltonian for the quantized laser field photons with the frequency ω_{23}. The second term is the Hamiltonian for the molecular levels with energies ϵ_i. The third term is for the pumping laser. The electric dipole coupling strength is given by μ and the pump laser has frequency ω_{14}. $E_p(t)$ allows for the potentially noisy modulations of the pump mechanism. The fourth term is for the coupling between the dye levels and the quantized field. The dipole transition coupling strength is g and this expression contains $(a+a^\dagger)$ rather than $(a-a^\dagger)$ for the electric dipole coupling because we assume standing waves in the laser cavity.[5(e)] The remaining terms are stochastic Hamiltonians representing the nonradiative level transitions (or radiative ones not concerning the pump and lasing processes) depicted in Fig. 1, and a cavity noise Hamiltonian, $\tilde{H}_c(t)$, to account for cavity relaxation. Note that each contribution to H is manifestly Hermitian.

Each stochastic contribution to the Hamiltonian is assumed to be Gaussian, to have zero mean, and to be statistically independent of each of the others. These facts may be expressed by

$$\langle \tilde{H}_{ij}(t)\rangle = 0, \quad \langle \tilde{H}_c(t)\rangle = 0 , \tag{6}$$

$$\langle \tilde{H}_{ij}(t)\tilde{H}_{kl}(s)\rangle = Q_{ijkl}\delta(t-s) , \tag{7}$$

$$\langle \tilde{H}_{ij}(t)\tilde{H}_c(t)\rangle = 0 , \tag{8}$$

with

$$Q_{ijkl} = Q_{ijkl}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \tag{9}$$

with

$$Q_{ijij} = \Gamma_{ij} .$$

The Dirac delta correlations in Eq. (7) represent the assumption that these quantities fluctuate on a very short time scale compared with laser field relaxation time scales.

Fox[9] has analyzed the effects of performing stochastic averages over the stochastic contributions of the Hamiltonian. The exact result of such averaging, given the assumptions of Gaussian white stochasticity in Eqs. (6)—(9), is given by (see Appendix A)

$$i\hbar\frac{\partial\rho}{\partial t} = [H_0 + H_I, \rho] + i\hbar R\rho + i\hbar D\rho \tag{10}$$

in which R and D are superoperators which cannot be expressed by commutators. The quantities in (10) are

$$H_0 = h\omega_{23}a^\dagger a + \sum_{i=1}^{4} |i\rangle\epsilon_i\langle i| , \tag{11}$$

$$H_I = \mu\cos(\omega_{14}t)(|4\rangle E_p(t)\langle 1| + |1\rangle E_p^*(t)\langle 4|) + g(a+a^\dagger)(|3\rangle\langle 2| + |2\rangle\langle 3|) , \tag{12}$$

$$R = R^{(3,4)} + R^{(1,4)} + R^{(2,3)} + R^{(1,2)} + R^{(1,3)} , \tag{13}$$

where

$$\begin{aligned} R^{(i,j)} = {} & \frac{-1}{\hbar^2}\Gamma_{ij}\frac{e^{-\beta\Delta E_{ij}}}{Z}|i\rangle\langle i|\cdot|i\rangle\langle i| - \frac{1}{\hbar^2}\frac{\Gamma_{ij}}{Z}|j\rangle\langle j|\cdot|j\rangle\langle j| \\ & + \frac{1}{\hbar^2}\frac{\Gamma_{ij}}{Z}|i\rangle\langle j|\cdot|j\rangle\langle i| + \frac{1}{\hbar^2}\Gamma_{ij}\frac{e^{-\beta\Delta E_{ij}}}{Z}|j\rangle\langle i|\cdot|i\rangle\langle j| \\ & - \frac{1}{2\hbar^2}\frac{\Gamma_{ij}}{Z}(1+e^{-\beta\Delta E_{ij}})(|i\rangle\langle i|\cdot|j\rangle\langle j| + |j\rangle\langle j|\cdot|i\rangle\langle i|) . \end{aligned} \tag{14}$$

In this expression, $\beta = 1/k_BT$ where k_B is Boltzmann's constant and T is the temperature; $\Delta E_{ij} = \epsilon_i - \epsilon_j$ and $Z = 1 + e^{-\beta\Delta E_{ij}}$. Because we will be dealing with a laser for which $\beta\Delta E_{ij} \gg 1$, we will neglect the terms involving $e^{-\beta\Delta E_{ij}}$. This amounts to neglecting upward transitions while keeping the downward transitions. In the context of Fig. 1, this means we keep the downward pointing transitions but not the upward pointing ones, which are suppressed by a Boltzmann factor in the exact expressions. Making this approximation, which is very good, means

$$\begin{aligned} R^{(i,j)} = {} & -\frac{1}{\hbar^2}\Gamma_{ij}|j\rangle\langle j|\cdot|j\rangle\langle j| + \frac{1}{\hbar^2}\Gamma_{ij}|i\rangle\langle j|\cdot|j\rangle\langle i| \\ & - \frac{1}{2\hbar^2}\Gamma_{ij}(|i\rangle\langle i|\cdot|j\rangle\langle j| + |j\rangle\langle j|\cdot|i\rangle\langle i| . \end{aligned} \tag{15}$$

In both Eqs. (14) and (15), the dots $\cdot$ indicate where to put the density matrix upon which these superoperators act. Finally, when the upward transitions are neglected, D is given by

$$D = \sum_n [-n\lambda|n\rangle\langle n|\cdot|n\rangle\langle n| + (n+1)\lambda|n\rangle\langle n+1|\cdot|n+1\rangle\langle n|] \tag{16}$$

in which $|n\rangle$ denotes the n-photon state of the laser field, and λ is the rate parameter for the decay of the field. The factors of n and $(n+1)$ preceding λ in Eq. (16) are the boson enhancement factors appropriate for photons. Notice that the $R^{(i,j)}$'s act exclusively on the dye levels whereas the D superoperator acts exclusively on the photon states. Moreover, the process of stochastic averaging did not in any way alter either H_0 or H_I in Eq. (10) as compared with Eq. (3). This is a consequence of the Dirac delta correlations in Eq. (7) and would not occur if the nonMarkovian correlations were assumed in Eq. (7) for these very fast processes. Equation (10) is still stochastic, even after all this averaging because it still contains the stochastic pump laser field, $E_p(t)$.

The next step of this analysis is to convert Eq. (10) into an interaction picture representation with respect to H_0. Define $\hat{\rho}$ by

$$\rho=\exp(-i\hbar^{-1}t[H_0,\cdot])\hat{\rho} \tag{17}$$

in which we exponentiate the commutator superoperator, $[H_0,\cdot]$. We can show straightforwardly that this yields

$$i\hbar\frac{\partial\rho}{\partial t}=[H_I(t),\hat{\rho}]+i\hbar R\hat{\rho}+i\hbar D\hat{\rho} \tag{18}$$

in which $H_I(t)$ is the interaction picture interaction Hamiltonian

$$H_I(t)=\mu(|4\rangle E_p(t)\langle 1|+|1\rangle E_p^*(t)\langle 4|)+g(|3\rangle a\langle 2|+|2\rangle a^\dagger\langle 3|) \tag{19}$$

in which only the energy conserving terms have been retained.[5] Notice that both the $R^{(i,j)}$'s and the D are unaltered by this step. For D, this is a consequence of the fact that $[H_0,\cdot]$ and D act in orthogonal portions of the direct product Hilbert space. For the $R^{(i,j)}$'s, it is a consequence of their particular structure and that of $[H_0,\cdot]$. Equation (18) may now be rendered as a closed system of eight operator density-matrix equations

$$i\hbar\frac{\partial\rho_{11}}{\partial t}=\mu[E_p^*(t)\hat{\rho}_{41}-E_p(t)\hat{\rho}_{14}]+\frac{i}{\hbar}(\Gamma_{12}\hat{\rho}_{22}+\Gamma_{13}\hat{\rho}_{33}+\Gamma_{14}\hat{\rho}_{44})+i\hbar D\hat{\rho}_{11}\ , \tag{20}$$

$$i\hbar\frac{\partial\rho_{22}}{\partial t}=g(a^\dagger\hat{\rho}_{32}-\hat{\rho}_{23}a)+\frac{i}{\hbar}(\Gamma_{23}\hat{\rho}_{33}-\Gamma_{12}\hat{\rho}_{22})+i\hbar D\hat{\rho}_{22}\ , \tag{21}$$

$$i\hbar\frac{\partial\rho_{33}}{\partial t}=g(a\hat{\rho}_{23}-\hat{\rho}_{32}a^\dagger)+\frac{i}{\hbar}(\Gamma_{34}\hat{\rho}_{44}-\Gamma_{23}\hat{\rho}_{33}-\Gamma_{13}\hat{\rho}_{33})+i\hbar D\hat{\rho}_{33}\ , \tag{22}$$

$$i\hbar\frac{\partial\rho_{44}}{\partial t}=\mu[E_p(t)\hat{\rho}_{14}-E_p^*(t)\hat{\rho}_{41}]+\frac{i}{\hbar}(-\Gamma_{34}\hat{\rho}_{44}-\Gamma_{14}\hat{\rho}_{44})+i\hbar D\hat{\rho}_{44}\ , \tag{23}$$

$$i\hbar\frac{\partial\rho_{23}}{\partial t}=g(a^\dagger\hat{\rho}_{33}-\hat{\rho}_{22}a^\dagger)-\frac{i}{2\hbar}\Gamma_{23}\hat{\rho}_{23}\ , \tag{24a}$$

$$i\hbar\frac{\partial\hat{\rho}_{32}}{\partial t}=g(a\hat{\rho}_{22}-\hat{\rho}_{33}a)-\frac{i}{2\hbar}\Gamma_{23}\hat{\rho}_{32}\ , \tag{24b}$$

$$i\hbar\frac{\partial\hat{\rho}_{14}}{\partial t}=\mu[E_p^*(t)\hat{\rho}_{44}-E_p^*(t)\hat{\rho}_{11}]-\frac{i}{2\hbar}\Gamma_{14}\hat{\rho}_{14}\ , \tag{25a}$$

$$i\hbar\frac{\partial\hat{\rho}_{41}}{\partial t}=\mu[E_p(t)\hat{\rho}_{11}-E_p(t)\hat{\rho}_{44}]-\frac{i}{2\hbar}\Gamma_{14}\hat{\rho}_{41}\ . \tag{25b}$$

Because these density-matrix elements with respect to the molecular levels are still operators in the photon Hilbert space, it is crucial to respect the ordering of a and $a^\dagger$ with respect to them.

The set of equations, (20)–(25), provides a detailed quantum-mechanical description of the stochastically pumped laser. In the following sections we will introduce approximations to obtain a more tractable, but still accurate, description of the laser.

III. ADIABATIC ELIMINATION OF OFF-DIAGONAL MATRIX ELEMENTS

The dynamics of the laser will be characterized by several time scales. The fastest processes are those which have just been averaged over, the level and cavity stochastic processes. The decay rates from level 4 to level 3 and level 2 to level 1 are the next fastest processes, which result in the populations of levels 4 and 2 being very small compared to those of levels 3 and 1. These relaxation rates are given by $(1/\hbar^2)\Gamma_{34}$ and $(1/\hbar^2)\Gamma_{12}$. Slower still are the relaxation rates $(1/\hbar^2)\Gamma_{14}$, $(1/\hbar^2)\Gamma_{23}$, and $(1/\hbar^2)\Gamma_{13}$. These may differ from $(1/\hbar^2)\Gamma_{34}$ and $(1/\hbar^2)\Gamma_{12}$ by one or more orders of magnitude. Usually, the slowest process involved is the cavity decay rate, λ. The precise values of these rates depends on the particular system being studied, but the ordering given above is quite common, and occurs in many different laser systems.[12]

Based on these premises, we may neglect Γ_{14} and the λ implicit in D compared with Γ_{34} in Eq. (23). This implies the "adiabatic elimination" of $\hat{\rho}_{44}$ by the identity

$$\hat{\rho}_{44}=\frac{\hbar\mu}{i\Gamma_{34}}[E_p(t)\hat{\rho}_{14}-E_p^*(t)\hat{\rho}_{41}]\ . \tag{26}$$

Anywhere else in Eqs. (20)–(25) in which $\hat{\rho}_{44}$ occurs without Γ_{34} it is to be neglected whereas terms of order $\Gamma_{34}\hat{\rho}_{44}$ are kept. Thus, Eq. (25) yields

$$\hat{\rho}_{14}(t)=-\frac{\mu}{i\hbar}\int_0^t ds\exp\left[-\frac{1}{2\hbar^2}\Gamma_{14}(t-s)\right]E_p^*(s)\hat{\rho}_{11}(s) \tag{27}$$

and its Hermitian adjoint for $\hat{\rho}_{41}$. We have assumed here that $\hat{\rho}_{14}(0)=0=\hat{\rho}_{41}(0)$. Similarly, we assume $\hat{\rho}_{23}(0)=0=\hat{\rho}_{32}(0)$ below.

Equation (27) may be used in Eqs. (20) and (26) to yield

$$\mu[E_p^*(t)\hat{\rho}_{41}(t)-E_p(t)\hat{\rho}_{14}(t)]=-\mu^2\frac{i}{\hbar}\int_0^t ds\exp\left[-\frac{\Gamma_{14}}{2\hbar^2}(t-s)\right]\times[E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]\hat{\rho}_{11}(s)\ . \tag{28}$$

It is convenient to introduce the abbreviations

$$\gamma_{ij}=\frac{\Gamma_{ij}}{\hbar^2},\quad \bar{\mu}=\frac{\mu}{\hbar},\quad \bar{g}=\frac{g}{\hbar}\ . \tag{29}$$

Equation (24) may be solved to yield

$$\hat{\rho}_{23}(t)=-i\bar{g}\int_0^t ds\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right]\times[a^\dagger\hat{\rho}_{33}(s)-\hat{\rho}_{22}(s)a^\dagger] \tag{30}$$

and its Hermitian adjoint.

Putting the consequences of Eqs. (26)–(30) together in Eqs. (20)–(25) provides the contracted dynamical description

$$\frac{\partial\hat\rho_{11}}{\partial t}=-\bar\mu^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]\hat\rho_{11}(s)+(\gamma_{12}\hat\rho_{22}+\gamma_{13}\hat\rho_{33})+D\hat\rho_{11}\;, \tag{31}$$

$$\frac{\partial\hat\rho_{22}}{\partial t}=-\gamma_{12}\hat\rho_{22}+\gamma_{23}\hat\rho_{33}+\bar g^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][a^\dagger\hat\rho_{33}(s)a-\hat\rho_{22}(s)a^\dagger a-a^\dagger a\hat\rho_{22}(s)+a^\dagger\hat\rho_{33}(s)a]+D\hat\rho_{22} \tag{32}$$

$$\frac{\partial\hat\rho_{33}}{\partial t}=-(\gamma_{13}+\gamma_{23})\hat\rho_{33}+\bar\mu^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]\hat\rho_{11}(s)$$
$$+\bar g^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][a\hat\rho_{22}(s)a^\dagger-\hat\rho_{33}(s)aa^\dagger-aa^\dagger\hat\rho_{33}(s)+a\hat\rho_{22}(s)a^\dagger]+D\hat\rho_{33}\;. \tag{33}$$

The photon density-matrix operator is not an independent quantity and is defined by $\hat\sigma=\hat\rho_{11}+\hat\rho_{22}+\hat\rho_{33}$ since we are neglecting $\hat\rho_{44}$ in this order of approximation. It satisfies the equation

$$\frac{\partial\hat\sigma}{\partial t}=D\hat\sigma+\bar g^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][2a^\dagger\hat\rho_{33}(s)a-\hat\rho_{33}(s)aa^\dagger-aa^\dagger\hat\rho_{33}(s)+2a\hat\rho_{22}(s)a^\dagger-\hat\rho_{22}(s)a^\dagger a-a^\dagger a\hat\rho_{22}(s)] \tag{34}$$

which depends directly on the solutions to Eqs. (31)—(33). Equations (31)—(33) provide a contracted alternative to Eqs. (20)—(25), and are a very good approximation to them if the time scales are well separated; this is true in many laser systems of interest, including the dye laser. In the following sections we will examine still further reductions of these equations; one of these reductions will provide us with the equivalent of a semiclassical theory, while the other will be a reduced density-matrix description of the quantized laser field.

IV. PHOTON STATE CONTRACTION

Up to now, we have described a quantized laser field interacting with a single four-level molecule. In this section, we will perform a photon state contraction of the description and obtain dynamical equations closely related to the phenomenological, semiclassical equations.[5] In addition, we generalize our treatment to N (four-level) molecules. It is to be noted that we obtain the electric field behavior directly from the density-matrix descriptions and we do not have to introduce an auxiliary account of the electric field and polarization as is usually done, e.g., in Louisell's account.[5(d)]

It is convenient to introduce the generalization to N molecules first, since the $N=1$ special case follows immediately. To treat N molecules, the Hamiltonian given in Eq. (6) must be enlarged to include terms for each molecule. The corresponding Hilbert space also must be enhanced by factoring in a finite dimensional Hilbert space for each molecule. The details are given in Appendix B. The result is that we obtain equations similar to Eqs. (31)—(34) except that Eqs. (31)—(33) are multiplied by an overall factor of N, whereas Eq. (34) has a factor of N multiplied into only the integral term in its right-hand side (rhs). The consequences of these factors of N are delineated in the following.

The number of molecules in state $|\,i\,\rangle$ is given by

$$\sum_n N\langle n\,|\hat\rho_{ii}|\,n\rangle\equiv N_i \quad \text{for } i=1,2,3 \tag{35}$$

where N_i is a number between 0 and N. Conservation of molecules implies

$$N_1+N_2+N_3=N=\text{const}\;. \tag{36}$$

The laser field intensity is the expectation value of the photon number operator, $a^\dagger a$, and is given by

$$\sum_n \langle n\,|a^\dagger a\hat\sigma|\,n\rangle\equiv I\;. \tag{37}$$

Equations (35) and (37) are consistent with the additional, but not independent, identity

$$\sum_n N\langle n\,|a^\dagger a\hat\rho_{ii}|\,n\rangle\equiv N_i I\;. \tag{38}$$

With these identities, the contraction of Eqs. (31)—(34), appropriately reinterpreted for N molecules, proceeds by tracing each equation over photon number expectation values. For example, Eq. (31) yields

$$\frac{dN_1}{dt}=-\bar\mu^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]N_1(s)+(\gamma_{12}N_2+\gamma_{13}N_3)\;. \tag{39}$$

The D term in Eq. (31) gives rise to zero when we use Eq. (16) for D

$$\sum_n N\langle n\,|D\hat\rho_{11}|\,n\rangle=N\left[\sum_{n=0}^{\infty}(-n\lambda)\langle n\,|\rho_{11}|\,n\rangle+\sum_{n=0}^{\infty}(n+1)\lambda\langle n+1\,|\hat\rho_{11}|\,n+1\rangle\right]=0\;. \tag{40}$$

The last step in Eq. (40) follows from shifting the summation index on the second sum. This cancellation will hold also for $\hat{\rho}_{22}$ and $\hat{\rho}_{33}$.

Inspection of Eqs. (31)—(34) justifies the conclusion that for $i=1,2,3$, $\hat{\rho}_{ii}$ is a function of the operator combination $a^\dagger a$ (or $aa^\dagger = a^\dagger a+1$) and not a function of the separate linear operators, a or $a^\dagger$. Therefore, it is convenient and valid to write $\hat{\rho}_{ii}(n)\equiv\langle n\,|\hat{\rho}_{ii}|\,n\rangle$, or $\hat{\rho}_{ii}(n+1)\equiv\langle n+1\,|\hat{\rho}_{ii}|\,n+1\rangle$. In Eqs. (32)—(34), we also meet combinations such as $a^\dagger a\hat{\rho}_{ii}$ and $a\hat{\rho}_{ii}a^\dagger$, as well as others. The ordering of these operators has been carefully preserved throughout the analysis, and it has the following consequences during the contraction process: Let $f=f(a^\dagger a)$ be any function of $a^\dagger a$, which is a continuous and infinitely differentiable function of its argument. We have the following identities which are easily proved:

$$\sum_n \langle n\,|f\,|\,n\rangle = \sum_n f(n)\,, \tag{41}$$

$$\sum_n \langle n\,|a^\dagger a f\,|\,n\rangle = \sum_n nf(n)\,, \tag{42}$$

$$\sum_n \langle n\,|a^\dagger f a\,|\,n\rangle = \sum_n nf(n-1)\,, \tag{43}$$

$$\sum_n \langle n\,|aa^\dagger f\,|\,n\rangle = \sum_n (n+1)f(n)\,, \tag{44}$$

$$\sum_n \langle n\,|afa^\dagger\,|\,n\rangle = \sum_n (n+1)f(n+1)\,. \tag{45}$$

Applied to Eq. (38), these results imply

$$\sum_n Nn\hat{\rho}_{ii}(n) = N_i I\,. \tag{46}$$

Whereas for the other kind of combination which occurs in Eq. (32), for example, $a^\dagger\hat{\rho}_{33}a$, they imply

$$\sum_n N\langle n\,|a^\dagger\hat{\rho}_{33}a\,|\,n\rangle = \sum_n Nn\hat{\rho}_{33}(n-1) = N_3 I + N_3\,. \tag{47}$$

Therefore, the contraction of Eq. (32) yields

$$\frac{d}{dt}N_2 = -\gamma_{12}N_2 + \gamma_{23}N_3 + 2\bar{g}^2 \times \int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right] \times [N_3(s)I(s) + N_3(s) - N_2(s)I(s)]\,. \tag{48}$$

The contraction of Eq. (33) yields

$$\frac{d}{dt}N_3 = -(\gamma_{13}+\gamma_{23})N_3 + \bar{\mu}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s) + E_p^*(t)E_p^*(s)]N_1(s) + 2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_2(s)I(s) - N_3(s) - N_3(s)I(s)]\,. \tag{49}$$

To obtain the intensity equation, Eq. (34) must be multiplied by $a^\dagger a$ and then traced over the photon states. Part of this particular contraction requires

$$\begin{aligned}\sum_{n=0}^{\infty}\langle n\,|a^\dagger a D\hat{\sigma}\,|\,n\rangle &= \sum_{n=0}^{\infty} n[-n\lambda\hat{\sigma}(n) + (n+1)\lambda\hat{\sigma}(n+1)] \\ &= \sum_{n=0}^{\infty}[-n^2\lambda\hat{\sigma}(n) + (n-1)n\lambda\hat{\sigma}(n)] = -\sum_{n=0}^{\infty}\lambda n\hat{\sigma}(n) = -\lambda I\end{aligned} \tag{50}$$

as follows from Eq. (37). Therefore, we get

$$\begin{aligned}\frac{d}{dt}I &= -\lambda I + 2\bar{g}^2 N\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right] \\ &\qquad\times\sum_n [n^2\hat{\rho}_{33}(n-1,s) - n(n+1)\hat{\rho}_{33}(n,s) + n(n+1)\hat{\rho}_{22}(n+1,s) - n^2\hat{\rho}_{22}(n,s)] \\ &= -\lambda I + 2\bar{g}^2 N\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right]\left[\sum_n [(n+1)\hat{\rho}_{33}(n,s) - n\hat{\rho}_{22}(n,s)]\right] \\ &= -\lambda I + 2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s) + N_3(s) - N_2(s)I(s)]\,.\end{aligned} \tag{51}$$

Notice the cancellation of all terms of order n^2 in Eq. (51).

So far, we have not made use of the fact that γ_{12} is a very fast rate. This fact can be used to adiabatically eliminate N_2 from further consideration. All terms in Eqs. (39), (49),and (51) of order N_2 can be neglected whereas terms of order $\gamma_{12}N_2$ must be retained and replaced by the adiabatic elimination identity which follows from Eq. (48)

$$\gamma_{12}N_2=\gamma_{23}N_3+2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s)+N_3(s)-N_2(s)I(s)] \tag{52}$$

in which the N_2 term on the right-hand side is self-consistently dropped. Putting Eq. (52) into Eq. (39) yields

$$\frac{d}{dt}N_1=(\gamma_{23}+\gamma_{13})N_3-\bar{\mu}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]N_1(s)$$
$$+2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s)+N_3(s)]\ . \tag{53}$$

Equation (49) becomes

$$\frac{d}{dt}N_3=-(\gamma_{13}+\gamma_{23})+\bar{\mu}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]N_1(s)$$
$$-2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s)+N_3(s)]\ . \tag{54}$$

Equation (51) becomes

$$\frac{d}{dt}I=-\lambda I+2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s)+N_3(s)]\ . \tag{55}$$

Clearly, to this order of approximation, we still preserve the total number of molecules, since $\dot{N}_1=-\dot{N}_3$. Therefore, it is correct to eliminate N_1 through

$$N_1=N-N_3\ . \tag{56}$$

In the next section we will proceed with the final reduction of these equations, which will be similar to well-known semiclassical equations. At this stage, however, it is useful to note the structure of our results.

Two different memory integrals occur in Eqs. (54) and (55). The memory integral in Eq. (54) for the pump terms involves $E_p(t)E_p^*(s)+E_p^*(t)E_p(s)$. If the pump field is stochastic, then the fluctuations must be incorporated into this structure. Only if the memory effect can be neglected in Eq. (54) can we approximate this structure by $2I_p(t)$, the pump intensity at a single time. Only then is the phenomenological treatment of pumping fluctuations used by earlier investigators partially justified.[2–4]

V. COMPARISON WITH SEMICLASSICAL THEORY AND NUMERICAL SIMULATION RESULTS

On performing the photon state contraction of the last section, we have finally obtained two coupled equations for the upper lasing level population and the laser intensity. They are

$$\frac{d}{dt}N_3=-(\gamma_{13}+\gamma_{23})N_3+\bar{\mu}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{14}}{2}(t-s)\right][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)][N-N_3(s)]$$
$$-2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s)+N_3(s)] \tag{57}$$

and

$$\frac{d}{dt}I=-\lambda I+2\bar{g}^2\int_0^t ds\,\exp\left[-\frac{\gamma_{23}}{2}(t-s)\right][N_3(s)I(s)+N_3(s)]\ . \tag{58}$$

These equations contain the effects of quantum fluctuations of the laser field (since we have traced over the photon number states). They also contain the effects of the pump stochasticity, and reveal several novel features on examination. The right-hand side of Eq. (57) shows clearly the dependence of N_3 (and hence I) on the fluctuating amplitude of the pump field $E_p(t)$. The exponentially decaying terms in the memory integrals contain relevant relaxations of the molecular levels. The relative time scales of the pump laser fluctuations and these decay rates will determine the eventual behavior of the integrals.

There are some special cases in which these equations reduce to forms which are well known.

(i) Constant pump intensity: In this case, the equations simply become, with $I_{po}=E_{po}E_{po}^*$

$$\frac{d}{dt}N_3=G-\lambda' N_3-BN_3(I+1)\ , \tag{59}$$

$$\frac{d}{dt}I=-\lambda I+BN_3(I+1)\ , \tag{60}$$

with

$$G=\frac{4\bar{\mu}^2}{\gamma_{14}}I_{po}N,\ \ \lambda'=(\gamma_{13}+\gamma_{23})+\frac{4\bar{\mu}^2}{\gamma_{14}}I_{po}$$
$$B=\frac{4\bar{g}^2}{\gamma_{23}}\ . \tag{61}$$

The assumption used in obtaining these equations is that

$$\int_0^t e^{-\gamma_{23}/2(t-s)}N_3(s)[I(s)+1]\simeq\frac{2}{\gamma_{23}}N_3(t)[I(t)+1] \tag{62}$$

and

$$\int_0^t e^{-\gamma_{23}/2(t-s)}N_3(s)ds\simeq\frac{2N_3(t)}{\gamma_{23}}\ . \tag{63}$$

Equations (59) and (60) are identical with the usual rate equations used to describe laser operation. If we now "adiabatically" eliminate N_3 through

$$N_3=\frac{G}{\lambda'+B(I+1)} \tag{64}$$

we obtain

$$\frac{d}{dt}I=-\lambda I+\frac{GB(I+1)}{\lambda'+B(I+1)}\ . \tag{65}$$

This equation, apart from the 1's in the parentheses, is identical to the (phenomenological) semiclassical equation given, for example, by Louisell in Ref. 5(d). The one's are a consequence of using a quantized formalism, which includes spontaneous emission.

(ii) If the pump field is not constant, but a very fast stochastic process, we may still obtain somewhat simplified equations which can be used to obtain numerical results. In addition to Eqs. (62) and (63) we have to approximate the pump noise in Eq. (57). Taking $E_p(t)$ to be simply of the form

$$E_p=\sqrt{I_{po}}+\xi(t)\ , \tag{66}$$

where $\xi(t)$ is Gaussian white noise, the pump term consists of four parts

$$2\bar{\mu}^2\int_0^t ds\, e^{-\gamma_{14}/2(t-s)}\{[\sqrt{I_{po}}+\xi(t)][\sqrt{I_{po}}+\xi(s)]\}[N-N_3(s)]$$
$$=2\bar{\mu}^2[N-N_3(t)]\int_0^t ds\, e^{-\gamma_{14}/2(t-s)}[I_{po}+\sqrt{I_{po}}\xi(s)+\sqrt{I_{po}}\xi(t)+\xi(s)\xi(t)] \tag{67}$$

in which N_3 has been treated as slowly varying compared to all other factors, which contain white noise. The first and third terms immediately give us

$$\frac{4\bar{\mu}^2}{\gamma_{14}}[N-N_3(t)]I_{po}$$

and

$$\frac{4\bar{\mu}^2}{\gamma_{14}}[N-N_3(t)]\xi(t)\sqrt{I_{po}}\ .$$

The last term may be approximated by $2\bar{\mu}^2\times[N-N_3(t)]\xi(t)^2$. A heuristic way of recognizing this is to realize that since the time scale for fluctuations of $\xi(t)$ is very short (compared to $1/\gamma_{14}$), only very small time differences will contribute to the integral, for which the exponential $e^{-\gamma_{14}/2(t-s)}$ is essentially unity.

The second term, $2\bar{\mu}^2[N-N_3(t)]\sqrt{I_{po}}\int_0^t e^{-\gamma_{14}/2(t-s)}\xi(s)ds$ will actually generate a colored noise term $f(t)$ with a time scale $(1/\gamma_{14})$. One can show that if

$$\langle\xi(t)\xi(t')\rangle=Q\delta(t-t') \tag{68}$$

then, with $f(t)=\int_0^t e^{-\gamma_{14}/2(t-s)}\xi(s)ds$, we have

$$\langle f(t)f(t')\rangle=\frac{2Q}{\gamma_{14}^2}\left[\frac{\gamma_{14}}{2}e^{-\gamma_{14}/2|t-t'|}\right]\ .$$

If $f(t)$ is still on a fast time scale compared to the time scale for field fluctuations set by $1/\lambda$, we may still treat this term as white noise, i.e.,

$$\lim_{\gamma_{14}\to\infty}\frac{\gamma_{14}}{2}e^{-\gamma_{14}/2|t-t'|}\to 2\delta(t-t')\ . \tag{69}$$

In other words,

$$\lim_{\gamma_{14}\to\infty}\langle f(t)f(t')\rangle\to\bar{Q}\delta(t-t') \tag{70}$$

with $\bar{Q}=4Q/\gamma_{14}^2$. To simulate Gaussian white noise, we use the Box-Müller algorithm to generate Gaussian random numbers x_1 and x_2 from uniformly distributed numbers γ_1 and γ_2,[13]

$$x_1=\sqrt{-2\ln\gamma_1}\cos(2\pi\gamma_2)\sqrt{Q\Delta t}\ , \tag{71}$$

$$x_2=\sqrt{-2\ln\gamma_1}\sin(2\pi\gamma_2)\sqrt{Q\Delta t}\ , \tag{72}$$

where Δt is the time step used to integrate the equations of motion. To simulate the noise term $f(t)$, Q would have to be replaced by $\bar{Q}$, which thus gives us the same contribution as the third term, each being $(4\bar{\mu}^2/\gamma_{14})\times[N-N_3(t)]\sqrt{I_{po}}x_1$.

With these approximations, one may now simulate the Eqs. (57) and (58) numerically on the computer and obtain correlation functions of the intensity of the laser field. The correlation function

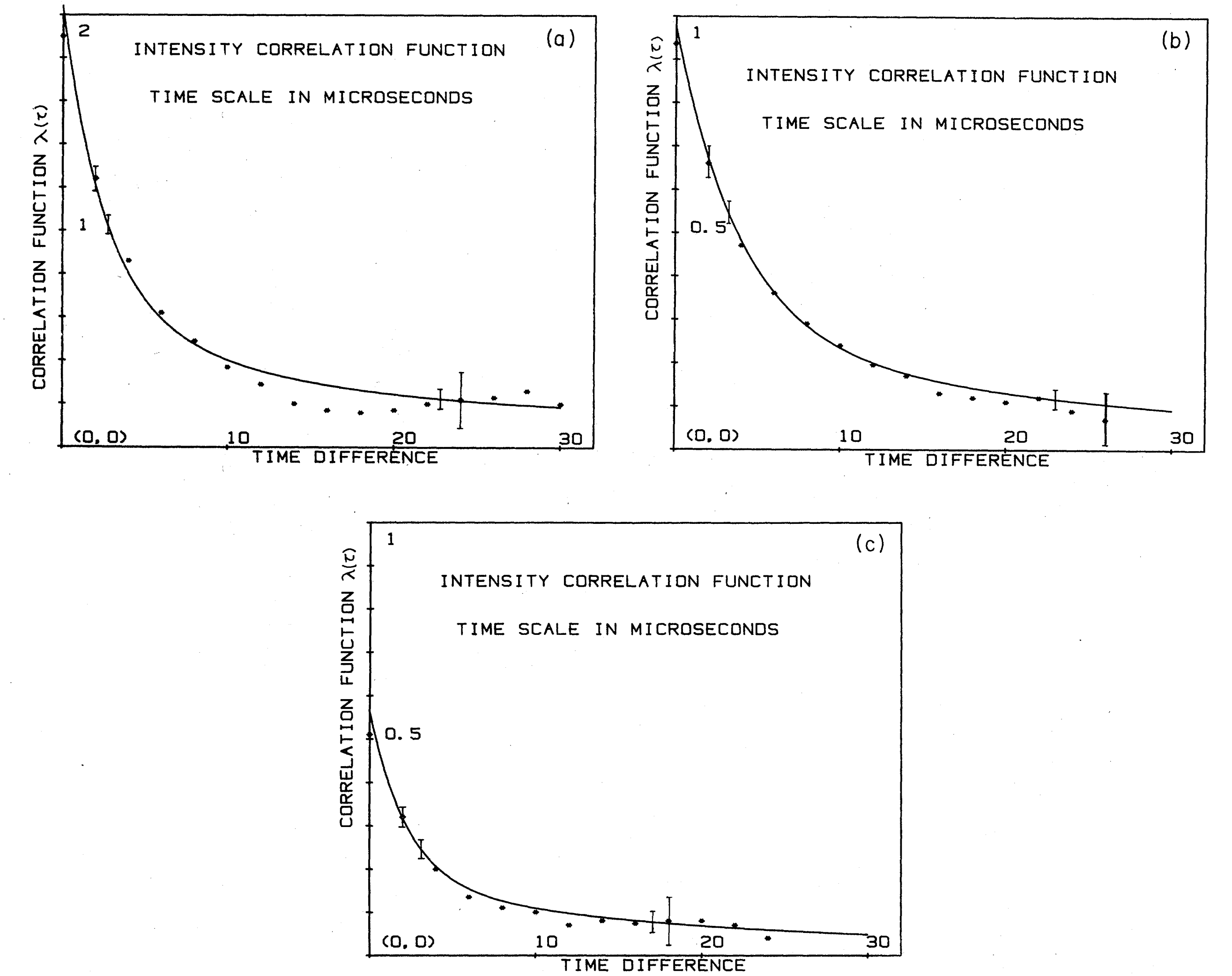

FIG. 2. (a), (b), (c): $\lambda(\tau)$ vs experimental data from Ref. 3; * are obtained from simulations. The error bars are estimated from the scatter of the experimental points and the results of the simulation for different realizations. (a) $[\lambda(0)]_{\rm expt}=2.04$; $I_{po}=6.0\times10^{8}$ $({\rm V/m})^2$; $B=0.0106\ {\rm sec}^{-1}$. (b) $[\lambda(0)]_{\rm expt}=0.98$; $I_{po}=2.0\times10^{9}$ $({\rm V/m})^2$; $B=0.0033\ {\rm sec}^{-1}$. (c) $[\lambda(0)]_{\rm expt}=0.57$; $I_{po}=2.4\times10^{9}$ $({\rm V/m})^2$; $B=0.0037\ {\rm sec}^{-1}$. The other parameters are common to all three figures $4\bar{\mu}^2/\gamma_{14}=10^{-4}\ {\rm sec}^{-1}$, $\gamma_{14}=1.4\times10^{5}\ {\rm sec}^{-1}$, $\lambda=5\times10^{5}\ {\rm sec}^{-1}$, $Q=7.0\times10^{2}$, $N=10^{10}$ molecules, $(\gamma_{13}+\gamma_{23})=10^{7}\ {\rm sec}^{-1}$, integration time step $=10^{-9}$ sec.

$$\lambda(\tau)=\frac{\langle \Delta I(t)\Delta I(t+\tau)\rangle}{\langle I\rangle^2} \tag{73}$$

may be calculated, suitable estimates being taken for the large number of dye parameters which occur in Eqs. (57)–(58). We have made such plausible estimates and obtained fits for the three intensity correlation functions which were measured experimentally.[3] The fits have been obtained by varying the average pump intensity I_{po} and the coefficient B, i.e., the three curves have been fit by varying two parameters. When the experiments reported in Refs. 3 and 7 were performed, I_{po} was not measured as an experimental parameter. At that time this was not considered a crucial measurement, since fits were being attempted with the one-parameter Haken-Risken theory of the laser. Also, when those measurements were performed, the pump intensity required for threshold varied by as much as 40–50% on different days, depending on the overlap of the pump and dye beams in the dye stream, the flow rate of the dye, and the overall alignment of the laser optics. However, the values of I_{po} used in our fits are of a reasonable magnitude for a low-loss laser cavity. The time step size taken was 1 ns and 300 realizations of 40 000 steps each were computed. Other parameters are stated in the figure captions, and the estimated errors in experimental and simulation results are shown in Figs. 2(a)–2(c). No "subtraction" procedures have been used,[2–4] which we regard as an important aspect of our theory.

VI. THE EFFECT OF QUANTUM FLUCTUATIONS

We have, until now, traced over the photon number states in order to show the correspondence of our equations with those of semiclassical laser theory. This procedure neglects the statistical features of the laser radiation which owe their origin to spontaneous emission, although it accounts for it in an averaged sense. In this section we will retain the stochastic effects due to both the pump laser and the intrinsic quantum fluctuations. The actual analysis of the equation which contains both these stochastic contributions is quite involved, and will be treated in detail elsewhere. In this section we merely outline the procedure to be followed.

Let us introduce the approximations represented by Eqs. (62) and (63) into Eqs. (33) and (34) for $\hat{\rho}_{33}$ and $\hat{\sigma}$, respectively. Neglect of the population of level 2 and the adiabatic elimination of $\hat{\rho}_{33}$ then gives

$$\dot{\hat{\sigma}}(n)=-\lambda n\sigma(n)+(n+1)\lambda\hat{\sigma}(n+1)+\frac{\bar{g}^2}{\gamma_{23}\left[\frac{\gamma_{13}^{+}\gamma_{23}}{2}\right]}\left[\frac{nG(t)\hat{\sigma}(n-1)}{1+\frac{\bar{g}^2 n}{\gamma_{23}\left[\frac{\gamma_{13}^{+}\gamma_{23}}{2}\right]}}-\frac{(n+1)G(t)\hat{\sigma}(n)}{1+\frac{\bar{g}^2(n+1)}{\gamma_{23}\left[\frac{\lambda_{13}^{+}\gamma_{23}}{2}\right]}}\right] . \quad (74)$$

Here

$$G(t)=\bar{\mu}^2 N\int_0^t ds\, e^{-\gamma_{14}/2(t-s)} \times[E_p(t)E_p^*(s)+E_p^*(t)E_p(s)] . \quad (75)$$

This equation has nearly the same form as the Scully-Lamb master equation.[5(e)] It can no longer be solved for steady state by putting the left-hand side to zero, since $G(t)$ is a time-dependent fluctuating quantity. In the special case that the pump field is constant, $G(t)$ is no longer stochastic and the Scully-Lamb equation is obtained as a limiting form.

It is possible to perform stochastic averages with respect to the pump fluctuations and obtain an approximate recursion equation for $\hat{\sigma}(n)$ in the steady state. This may be done by using the cumulant method of Fox[9] and leads to an equation of the form

$$\lambda(n+1)\hat{\sigma}(n+1)-\alpha(n)\hat{\sigma}(n)+\beta(n)\hat{\sigma}(n-1)=0 \quad (76)$$

which can be solved asymptotically for large n to obtain the form of the distribution $\hat{\sigma}(n)$. Analytic and numerical results on this equation will be presented in a forthcoming paper. This technique allows us to retain the statistical effect of quantum spontaneous emission and also stochastic pump effects. Our theoretical results for the photon number distribution will be compared with the recent experimental results of Mandel and co-workers[14] and also to the results obtained by simulating the phenomenological equation

$$\dot{E}=[a(t)-A\,|E\,|^2]E+f(t) ,$$

which contains both additive and multiplicative noise.

VII. CONCLUSION

Till now, the problem of pump noise in lasers was dealt with in a very *ad hoc* manner, by assuming a fluctuating pump parameter, which was either δ correlated or colored noise. We have formulated from first principles the interaction of a pump laser with the molecules of the secondary laser system. The pump laser is taken to be an intense, semiclassical field, while the laser field is quantized. Though a particular form for the stochasticity of the pump field still has to be assumed, the manner in which this noise appears even in the reduced semiclassical equations is seen to be different than has been assumed to date. We have, in particular, taken white noise for the pump laser, shown that this will produce colored noise in the secondary laser system, and fit some experimentally measured correlation functions for a single mode dye laser. An equation which includes quantum fluctuations has also been derived, using our basic equations in which the secondary laser field is quantized. This equation will be analyzed in a future publication.

APPENDIX A

The density matrix provides an alternative to the Schrödinger equation

$$i\hbar\frac{\partial}{\partial t}\psi=H\psi \quad (A1)$$

in the form

$$i\hbar\frac{\partial}{\partial t}\rho=[H,\rho] \quad (A2)$$

H is the Hamiltonian. The right-hand side of (A2) denotes the commutator of H and

$$[H,\rho]=H\rho-\rho H \quad (A3)$$

in which ρ is the density "matrix," or operator," which is related to ψ by

$$\rho=|\psi\rangle\langle\psi| . \quad (A4)$$

In (A4), $|\psi\rangle$ denotes the Schrödinger wave function, ψ, in Dirac's ket notation.

1. Operator calculus

To solve (A2), it is convenient to introduce the commutator operator, $[H,\cdot]$, defined by

$$[H,\cdot]M=HM-MH \quad (A5)$$

in which M is an arbitrary operator. While M acts in the original Hilbert space, $[H,\cdot]$ acts in the space of operators. It is called a "superoperator." The solution to (A2) can be written

$$\rho(t)=\exp\left[-\frac{i}{\hbar}t[H,\cdot]\right]\rho(0) \tag{A6}$$

in which $\rho(0)$ is the initial value density-matrix (operator), and the exponentiated superoperator is itself a superoperator which acts on $\rho(0)$. An easily proved identity, for arbitrary M, is

$$\exp\left[-\frac{i}{\hbar}t[H,\cdot]\right]M=\exp\left[-\frac{i}{\hbar}tH\right]M\exp\left[\frac{i}{h}tH\right]. \tag{A7}$$

Let $H=H_0+\tilde{H}(t)$ in which $\tilde{H}(t)$ is stochastic white noise and $[H_0,\tilde{H}(t)]\neq 0$

$$\rho(t)\equiv\exp\left[-\frac{i}{\hbar}t[H_0,\cdot]\right]\hat{\rho}(t). \tag{A8}$$

This yields an equation for $\hat{\rho}(t)$

$$i\hbar\frac{\partial}{\partial t}\hat{\rho}(t)=[H_I(t),\cdot]\hat{\rho}(t) \tag{A9}$$

in which $H_I(t)$ is defined by

$$H_I(t)=\exp\left[\frac{i}{\hbar}tH_0\right]\tilde{H}(t)\exp\left[-\frac{i}{\hbar}tH_0\right]. \tag{A10}$$

The solution to (A9) is not as easy to express as was the solution, (A6), to A(2) because of the t dependence in $H_I(t)$. For two different times, t and s, we find

$$[H_i(t),H_I(s)]\neq 0 \tag{A11}$$

because $[H_0,\tilde{H}(t)]\neq 0$.

We may overcome the commutativity problem expressed in (A11) by introducing the "t-ordered exponential." Consider the general vector equation

$$\frac{d}{dt}a=M(t)a \tag{A12}$$

in which a is an N-component vector and $M(t)$ is a t-dependent operator ($N\times N$ matrix) acting in the vector space in which a is found. The solution to (A12), with initial value vector a_0, is

$$\begin{aligned}a(t)&=a_0+\int_0^t ds\,M(s)a_0+\int_0^t ds_1\int_0^{s_1}ds_2M(s_1)M(s_2)a_0+\cdots\\&\quad+\int_0^t ds_1\int_0^{s_1}ds_2\cdots\int_0^{s_{n-1}}ds_nM(s_1)M(s_2)\cdots M(s_n)a_0+\cdots\\&=\left[1+\sum_{n=1}^{\infty}\int_0^t ds_1\int_0^{s_1}ds_2\cdots\int_0^{s_{n-1}}ds_nM(s_1)M(s_2)\cdots M(s_n)\right]a_0\equiv\underleftarrow{T}\exp\left[\int_0^t ds\,M(s)\right]a_0.\end{aligned} \tag{A13}$$

The third equality defines the symbol $\underleftarrow{T}\exp[\int_0^t ds\,M(s)]$, which must not be confused with the ordinary exponential, $\exp[\int_0^t ds\,M(s)]$.

We may now write the solution to (A9) in the form

$$\hat{\rho}(t)=\underleftarrow{T}\exp\left[-\frac{i}{\hbar}\int_0^t ds[H_I(s),\ ]\right]\hat{\rho}(0). \tag{A14}$$

For arbitrary $M(s)$, the analog of (A7) may be proved

$$\begin{aligned}\underleftarrow{T}\exp\left[-\frac{i}{\hbar}\int_0^t ds[H_I(s),\cdot]\right]M&=\underleftarrow{T}\exp\left[-\frac{i}{\hbar}\int_0^t dx\,H_I(s)\right]M\\&\quad\times\underrightarrow{T}\exp\left[\frac{i}{\hbar}\int_0^t ds\,H_I(s)\right].\end{aligned} \tag{A15}$$

The reduced density matrix is defined by

$$\rho_{\rm red}(t)=\exp\left[-\frac{i}{\hbar}tH_0\right]\langle\hat{\rho}(t)\rangle\exp\left[\frac{i}{\hbar}tH_0\right]. \tag{A16}$$

2. Cumulants and stochasticity

Equation (A16) does not provide a closed description for $\rho_{\rm red}(t)$ in terms of itself only, but depends upon the full density matrix, $\hat{\rho}(t)$. The idea of "contraction of the description" is to get a closed description. This means that we must find a closed description for $\rho_{\rm red}(t)$. The technique for achieving this goal is exhibited below.

Define the evolution superoperator, $E(t)$, which acts on operators in the Hilbert space, by

$$E(t)\equiv\left\langle\underleftarrow{T}\exp\left[-\frac{i}{\hbar}\int_0^t ds[H_I(s),\cdot]\right]\right\rangle. \tag{A17}$$

Therefore, we can write (A16) as

$$\rho_{\rm red}(t)=\exp\left[-\frac{i}{\hbar}tH_0\right][E(t)\hat{\rho}(0)]\exp\left[\frac{i}{\hbar}tH_0\right]. \tag{A18}$$

From this form of $\rho_{\rm red}(t)$, it follows that

$$i\hbar\frac{\partial}{\partial t}\rho_{\rm red}(t)=[H_0,\rho_{\rm red}(t)]+i\hbar\exp\left[-\frac{i}{\hbar}tH_0\right]\left[\left[\frac{\partial}{\partial t}E(t)\right]E^{-1}(t)E(t)\hat{\rho}(0)\right]\exp\left[\frac{i}{\hbar}tH_0\right]. \tag{A19}$$

It will be shown below that the superoperator combination $[\partial/\partial t E(t)]E^{-1}(t)$ can be written

$$\left[\frac{\partial}{\partial t}E(t)\right]E^{-1}(t)\equiv G(t) . \tag{A20}$$

Using (A7) twice, and (A18) once with (A20), permits us to rewrite (A19) as

$$i\hbar\frac{\partial}{\partial t}\rho_{\rm red}(t)=[H_0,\cdot]\rho_{\rm red}(t)+i\hbar\exp\left[-\frac{i}{\hbar}t[H_0,\cdot]\right]G(t)\exp\left[\frac{i}{\hbar}t[H_0,\cdot]\right]\rho_{\rm red}(t) . \tag{A21}$$

This is now a manifestly closed equation for the reduced density-matrix (operator) $\rho_{\rm red}(t)$. The first term on the right-hand side of (A21) is the only term present if there is no stochastic Hamiltonian. The second term involves a succession of three superoperators and is explicitly t dependent. This t dependence is the price paid in order to achieve the contraction of the description.

When $H_I\neq 0$, $G(t)$ does not vanish, and it is not expressible in the form of a simple commutator operator (superoperator), i.e.,

$$\exp\left[-\frac{i}{\hbar}t[H_0,\cdot]\right]G(t)\exp\left[\frac{i}{\hbar}t[H_0,\cdot]\right]\neq[H_{\rm eff}(t),\cdot] . \tag{A22}$$

Therefore, (A21) is not a special case of (A2), nor is it equivalent to (A1), with an effective Hamiltonian.

The operator cumulant expansion is expressed

$$\left\langle \underset{\leftarrow}{T}\exp\left[-\frac{i}{\hbar}\int_0^t ds\,[H_I(s),\cdot]\right]\right\rangle = \underset{\leftarrow}{T}\exp\left[\sum_{n=1}^{\infty}\int_0^t ds\,G^{(n)}(s)\right] . \tag{A23}$$

Explicit expressions exist for the nth operator cumulant, $G^{(n)}$, in terms of the moments of H_I. Comparison with (A20) verifies

$$G(t)=\sum_{n=1}^{\infty}G^{(n)}(t) . \tag{A24}$$

The first two cumulants are

$$G^{(1)}(t)=\left\langle -\frac{i}{\hbar}[H_I(t),\cdot]\right\rangle \tag{A25}$$

and

$$G^{(2)}(t)=-\frac{1}{\hbar^2}\int_0^t ds\,\langle [H_I(t),\cdot][H_I(s),\cdot]\rangle - \int_0^t ds\,G^{(1)}(t)G^{(1)}(s) . \tag{A26}$$

Equation (10) follows from these formulas.

APPENDIX B

The effect of having N molecules instead of only one shows up in the analogs to Eqs. (18) and (19)

$$i\hbar\frac{\partial\hat{\rho}}{\partial t}=[H_I(t),\hat{\rho}]+i\hbar\sum_{l=1}^{N}R_l\hat{\rho}+i\hbar D\hat{\rho} \tag{B1}$$

in which R_l corresponds with the definitions in Eqs. (13) and (15) except that the molecular states in these equations are now for the lth molecule; and $H_I(t)$ is given by

$$H_I(t)=\sum_{l=1}^{N}H_{I,l}(t)=\sum_{l=1}^{N}[\mu(|4_l\rangle E(t)\langle 1_l|+|1_l\rangle E_p^*(t)\langle 4_l|)+g(|3_l\rangle a\langle 2_l|+|2_l\rangle a^\dagger\langle 3_l|)] , \tag{B2}$$

where again the molecular states with subscript l are for the lth molecule. Our notation ignores factors of identity operators for each orthogonal factor of the Hilbert space, just as we earlier discussed for Eq. (5). Thus, $|3_l\rangle a\langle 2_l|$ is really $\prod_{k\neq l}^{N}|3_l\rangle a\langle 2_l|\times 1_k$. The correct eigenkets contain N indices: $|i_1 i_2\cdots i_N\rangle$, where $i_l=1,2,3,4$ for $l=1,2,\ldots,N$. The analogs to the diagonal equations (20) through (23) are

$$i\hbar\frac{\partial}{\partial t}\hat{\rho}_{i_1i_1i_2i_2\cdots i_Ni_N}=\sum_{l=1}^{N}\langle i_1i_2\cdots i_N|[H_{I,l}(t),\hat{\rho}]|i_1i_2\cdots i_N\rangle + i\hbar\sum_{l=1}^{N}\langle i_1i_2\cdots i_N|R_l\hat{\rho}|i_1i_2\cdots i_N\rangle+i\hbar D\hat{\rho}_{i_1i_1i_2i_2\cdots i_Ni_N} . \tag{B3}$$

This amounts to 4^N diagonal equations. The photon density matrix, which for one molecule was given by $\hat{\sigma}=\sum_{i=1}^{4}\hat{\rho}_{ii}$ [Eq. (34) wherein we have already adiabatically removed $\hat{\rho}_{44}$] is now given by the N-fold trace

$$\hat{\sigma}=\sum_{i_1=1}^{4}\sum_{i_2=1}^{4}\cdots\sum_{i_N=1}^{4}\hat{\rho}_{i_1i_1i_2i_2\cdots i_Ni_N} . \tag{B4}$$

The N-fold trace over $\hat{\rho}$ satisfies the N-molecule normalization requirement

$$\sum_{n=0}^{\infty}\sum_{i_1=1}^{4}\sum_{i_2=1}^{4}\cdots\sum_{i_N=1}^{4}\hat{\rho}_{i_1i_1i_2i_2\cdots i_Ni_N}=N . \tag{B5}$$

To obtain the analogs of Eqs. (20)—(25), we perform reductions of the density-matrix expressions such as in Eq. (B3). This is done by tracing over molecules 2 through N. Without loss of generality, we are isolating molecule 1 since the molecules are identical. Thus define $\hat{\rho}_{i_1 i_1}$ or $\hat{\rho}_{i_1 j_1}$ by

$$\hat{\rho}_{i_1 j_1} = \sum_{i_2=1}^{4} \cdots \sum_{i_N=1}^{4} \hat{\rho}_{i_1 j_1 i_2 i_2 \cdots i_N i_N} . \tag{B6}$$

This yields the reduced equations

$$i\hbar \frac{\partial \hat{\rho}_{i_1 j_1}}{\partial t} = \langle i_1 | [H_{I,1}(t), \hat{\rho}] | j_1 \rangle + i\hbar \langle i_1 | R_1 \hat{\rho} | j_1 \rangle + i\hbar D \hat{\rho}_{i_1 j_1} \delta_{i_1 j_1} + \delta_{i_1 j_1} g \sum_{l=2}^{N} ([a^\dagger, \hat{\rho}_{3_l 2_l}] + [a, \hat{\rho}_{2_l 3_l}]) \tag{B7}$$

in which $\delta_{i_1 j_1}$ is the Kronecker delta symbol. The first three terms of the right-hand side coincide with Eqs. (20)–(25). The extra terms, which only contribute to diagonal elements, result from the noncommutativity of the $\hat{\rho}$ molecular matrix element with the photon operators. It also follows immediately from Eq. (B7) that the $\hat{\sigma}$ defined in Eq. (B4) satisfies

$$i\hbar \frac{\partial \hat{\sigma}}{\partial t} = ihD\hat{\sigma} + g \sum_{l=1}^{N} ([a^\dagger, \hat{\rho}_{3_l 2_l}] + [a, \hat{\rho}_{2_l 3_l}]) . \tag{B8}$$

Adiabatic elimination of off-diagonal elements from the system of equations given by Eqs. (B7) and (B8) will result in the N-molecule analogs of Eqs. (31)–(33).

[1]Y. K. Wang and W. E. Lamb, Jr., Phys. Rev. A **8**, 873 (1973).

[2]R. Graham, M. Höhnerbach, and A. Schenzle, Phys. Rev. Lett. **48**, 1396 (1982).

[3]R. Short, L. Mandel, and R. Roy, Phys. Rev. Lett. **49**, 647 (1982).

[4]S. N. Dixit and P. Sahni, Phys. Rev. Lett. **50**, 1273 (1982).

[5](a) H. Risken, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1970); (b) H. Haken, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1970), Vol. XXV/2c; (c) M. Lax, in *Brandeis University Summer Institute Lectures*, edited by M. Chretien, E. P. Gross, and S. Deser (Gordon and Breach, New York, 1967), Vol. II; (d) W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973); (e) M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, Mass., 1974).

[6](a) A. Smith and J. A. Armstrong, Phys. Rev. Lett. **16**, 1169 (1966); (b) Phys. Lett. **19**, 650 (1966); (c) F. T. Arecchi, Phys. Rev. Lett. **15**, 912 (1966); (d) F. T. Arecchi, A. Berne, and P. Burlamacchi, *ibid.* **16**, 30 (1966); (e) F. T. Arecchi, V. De Giorgio, and B. Querzola, *ibid.* **19**, 1168 (1967); (f) F. T. Arecchi, G. S. Rodari, and A. Sona, Phys. Lett. **25A**, 59 (1967); (g) C. Freed and H. A. Haus, Phys. Rev. Lett. **15**, 943 (1965); (h) Phys. Rev. **141**, 287 (1966); (i) R. F. Chang, V. Korenman, C. O. Alley, and R. W. Detenbeck, *ibid.* **178**, 612 (1969); (j) F. Davidson and L. Mandel, Phys. Lett. **25A**, 700 (1967); (k) J. Appl. Phys. **39**, 62 (1968); (l) F. Davidson, Phys. Rev. **185**, 446 (1969); (m) D. Meltzer and L. Mandel, IEEE J. Quantum Electron. **QE-6**, 661 (1970); (n) D. Meltzer and L. Mandel, Phys. Rev. A **3**, 1763 (1971); (o) S. Chopra and L. Mandel, Phys. Rev. Lett. **30**, 60 (1973).

[7]K. Kaminishi, R. Roy, R. Short, and L. Mandel, Phys. Rev. A **24**, 370 (1981).

[8]R. B. Schaefer and C. R. Willis, Phys. Rev. A **13**, 1874 (1976).

[9]R. F. Fox, Phys. Rep. **48**, 179 (1978).

[10](a) A. Schenzle and H. Brand, Phys. Rev. A **20**, 1628 (1979); (b) R. Graham and A. Schenzle, *ibid.* **25**, 1731 (1982).

[11]R. F. Fox, Phys. Lett. **94A**, 281 (1983).

[12]See, for example, O. Svelto, *Principles of Lasers* (Plenum, New York, 1982), 2nd ed., Chap. 6.

[13]D. E. Knuth, *The Art of Computer Programming*, 2nd ed. (Addison-Wesley, Reading, Mass., 1981), Vol. II.

[14]P. Lett, R. Short, and L. Mandel, Phys. Rev. Lett. **52**, 341 (1984).