Photon distributions of lasers with first-order phase-transition analogies

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The photon-number distributions for a laser with a saturable-absorber and a dye laser are obtained by extensions of the Scully and Lamb optical-maser theory. The results are shown to be in correspondence with those of Lugiato *et al.* for the saturable-absorber laser and with those of Schaefer and Willis for the dye laser. Moreover, they are in a form which clearly shows the relation between the saturable-absorber laser and the dye laser. The thermodynamic potentials for the first-order phase-transition analogy are calculated and are found to be of the form predicted by Scott, Sargent, and Cantrell using semiclassical equations.

I. INTRODUCTION

Many systems exhibit transitions from disordered to ordered (or vice versa) behavior. Some of these are in thermal equilibrium; others are far from it when the transition occurs.[1,2] The laser falls in the second category. It is well understood from a microscopic viewpoint; its experimentally observed properties have been explained in some detail and predictions concerning its behavior have been verified.

To view the laser as a non-thermal-equilibrium system in the steady state, the pumping source can be regarded as a high-temperature reservoir and the laser's surroundings as a low-temperature reservoir (to which it loses energy). This analogy has been discussed in Refs. 2–4, and is displayed in the table presented by Scully in Refs. 2(d) and 4.

The laser model usually chosen for analysis[1,3] is one which leads directly to the similarity with a second-order phase transition in equilibrium systems. In thermodynamic language a phase transition is first order if the first derivatives of the Gibbs potential change discontinuously from one phase to the other. They are $S=-(\partial G/\partial T)_P$ and $V=(\partial G/\partial P)_T$, or their analogs in the case of the laser. In a second-order transition these quantities change continuously at the transition temperature, but the second derivatives show discontinuous changes.

It has been known for some time now that there are lasers which show first-order phase-transition analogies.[5–10] Two such systems have been studied: (i) the laser with a saturable absorber and (ii) the dye laser. The laser with an externally injected signal[2(a)11(a)] was also thought to be a system displaying a first-order phase-transition analogy, but the recent work of Lugiato[11(b)] shows that this is not true.

Semiclassical analyses have been performed for the laser with a saturable absorber[6,7,10] (which will be referred to as LSA in the future) as well as the dye laser,[9] which explain the hysteresis effects observed in the former[16] and predict similar effects for the dye laser. Quantum-mechanical calculations or semiclassical calculations in which Langevin noise sources are added have also been carried out which predict the photon statistics of the LSA[6,7,10] and the dye laser.[8,9] In this paper we wish to examine the photon distributions for both these lasers using extensions of the Scully-Lamb theory of the optical maser. For the LSA we do not present the theory in detail since the results can be obtained by simple generalizations of the theory given in Refs. 1. The calculations on the dye laser are more detailed; this is because intersystem crossing rates connect the singlet and triplet states, resulting in a system more complicated than the LSA. We obtain results in a form which clearly demonstrates the similarity between the two and our results are shown to be in correspondence to those of Lugiato *et al.* for the LSA and of Schaefer and Willis for the dye laser. The thermodynamic potentials are calculated, and shown to be of just the form predicted by Scott *et al.*[5] on the basis of the phase-transition analogy and the semiclassical equation for the field.

II. LASER WITH A SATURABLE ABSORBER

We give an outline of the experimental setup in Fig. 1. The pressure and discharge current in each tube can be independently controlled. This is required for the laser to operate in the region of interest—the bistable region.[6,16] A theoretical analysis will now be given, and we will find the photon-number distribution for the laser.

The active medium and the absorber are both

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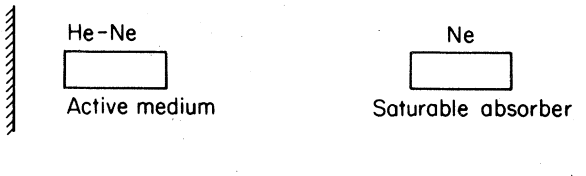

FIG. 1. Gas laser with a saturable absorber. (The plasma tubes usually have Brewster windows and the mirrors need not be flat.)

represented by homogeneously broadened two-level atoms (Fig. 2). Here r_a, r_b, r_c, and r_d are the rates at which atoms are introduced into levels a, b, c, and d, respectively. To produce gain in the active medium, r_a is larger than r_b, but r_d is greater than r_c in the absorber. The levels a, b, c, and d decay to e, f, g, and h at the rates γ_a, γ_b, γ_c, and γ_d. The linear cavity losses are simulated by another set of atoms with levels 1 and 2; only terms linear in the photon number are retained.

We will not repeat, but merely outline, the steps leading to the equation of motion for the matrix elements of the radiation density matrix. These are identical with those of Scully, Kim, and Lamb,[1(b)] except that we have now introduced an extra species of atom with levels c and d. The Hamiltonian for the system is, in the notation of Refs. 1(a) and 1(b),

$$\mathcal{H} = \hbar H$$

where

$$H = \nu a^\dagger a + (W_a \sigma_1^\dagger \sigma_1 + W_b \sigma_1 \sigma_1^\dagger) + (W_c \sigma_2^\dagger \sigma_2 + W_d \sigma_2 \sigma_2^\dagger) + g_1(a^\dagger \sigma_1 + a\sigma_1^\dagger) + g_2(a^\dagger \sigma_2 + a\sigma_2^\dagger) . \tag{2.1}$$

Here $\hbar\nu a^\dagger a$ is the field energy, ν being the laser frequency, while $a^\dagger$ and a are the creation and annihilation operators for the single-mode electromagnetic field. The energies of the atomic levels a, b, c, and d are $\hbar W_a$, $\hbar W_b$, $\hbar W_c$, and $\hbar W_d$. The σ's are raising and lowering operators for the atomic states of the active medium (subscript 1) and the absorber (subscript 2). The last two terms describe the interaction between the field and the atoms, where $g_1 = ex_{ab}\mathcal{E}/\sqrt{2}\hbar$ (a frequency) and $g_2 = ex_{cd}\mathcal{E}/\sqrt{2}\hbar$ are the coupling constants. The x's are dipole moments for the transitions. $\mathcal{E} = (\hbar\Omega/LA\epsilon_0)^{1/2}$ has the dimensions of an electric field, with Ω the free-field frequency, and L and A are the length and cross sectional area of the cavity.

Our aim is to obtain the equations of motion for the elements of the radiation- density matrix ρ. The procedure followed is straightforward; one first finds the change produced in the reduced (field) density-matrix elements by introducing an atom in the atomic level a, b, c, or d. This is found in terms of the decay rates of the levels, the detuning of the laser frequency from the atomic transition frequency and the numbers n and n' of the Fock states of the radiation field for which the matrix element is taken. An average, or coarse-grained rate of change of the field- density-matrix elements is then found by multiplying the change due to a single atom introduced in a given state by the rate of introduction of the atoms in that state and adding all these terms. Cavity losses are simulated by introducing an interaction with another hypothetical set of atoms and retaining only the terms linear in the photon number. This entire process is detailed in Refs. 1(a) and 1(b). Also, a similar process will be presented in some detail in the section on the dye laser. Here we just give the final coarse-grained equation of motion for the elements $\rho_{nn'}$ of the radiation-field density matrix. (The asterisks denote complex conjugate quantities.)

$$\begin{aligned}
\dot{\rho}_{nn'} = &-[(n+1)R_{1n,n'} + (n'+1)R^*_{1n',n}]\rho_{nn'}(t) + (R_{1n-1,n'-1} + R^*_{1n'-1,n-1})(nn')^{1/2}\rho_{n-1,n'-1}(t) \\
&+ (R'_{1n,n'} + R'^*_{1n',n})[(n+1)(n'+1)]^{1/2}\rho_{n+1,n'+1}(t) - (nR'_{1n-1,n'-1} + n'R'^*_{1n'-1,n-1})\rho_{nn'}(t) \\
&- [(n+1)R_{2n,n'} + (n'+1)R^*_{2n',n}]\rho_{nn'}(t) + (R_{2n-1,n'-1} + R^*_{2n'-1,n-1})(nn')^{1/2}\rho_{n-1,n'-1}(t) \\
&- (nR'_{2n-1,n'-1} + n'R'^*_{2n'-1,n-1})\rho_{nn'}(t) + (R'_{2n,n'} + R'^*_{2n',n})[(n+1)(n'+1)]^{1/2}\rho_{n+1,n'+1}(t) \\
&- \tfrac{1}{2}C(n+n')\rho_{nn'}(t) + C[(n+1)(n'+1)]^{1/2}\rho_{n+1,n'+1}(t) - \tfrac{1}{2}(n_2/n_1)C(n+1+n'+1)\rho_{nn'}(t) \\
&+ (n_2/n_1)C(nn')^{1/2}\rho_{n-1,n'-1}(t) .
\end{aligned} \tag{2.2}$$

The quantities contained in Eq. (2.2) are defined below:

$$R_{1n,n'} = r_a g_1^2 \left(\frac{\gamma_b(\gamma_{ab} + i\Delta_1) + g_1^2(n-n')}{\gamma_a\gamma_b(\gamma_{ab}^2 + \Delta_1^2) + 2\gamma_{ab}^2 g_1^2(n+1+n'+1) + g_1^2(n'-n)[g_1^2(n'-n) + i\Delta_1(\gamma_a - \gamma_b)]} \right), \tag{2.3}$$

$$R'_{1n,n'}=r_b g_1^2\left(\frac{\gamma_a(\gamma_{ab}-i\Delta_1)+g_1^2(n-n')}{\gamma_a\gamma_b(\gamma_{ab}^2+\Delta_1^2)+2\gamma_{ab}^2 g_1^2(n+1+n'+1)+g_1^2(n'-n)[g_1^2(n'-n)+i\Delta_1(\gamma_a-\gamma_b)]}\right), \tag{2.4}$$

$$R_{2n,n'}=r_c g_2^2\left(\frac{\gamma_d(\gamma_{cd}+i\Delta_2)+g_2^2(n-n')}{\gamma_c\gamma_d(\gamma_{cd}^2+\Delta_2^2)+2\gamma_{cd}^2 g_2^2(n+1+n'+1)+g_2^2(n'-n)[g_2^2(n'-n)+i\Delta_2(\gamma_c-\gamma_d)]}\right), \tag{2.5}$$

$$R'_{2n,n'}=r_d g_2^2\left(\frac{\gamma_c(\gamma_{cd}-i\Delta_2)+g_2^2(n-n')}{\gamma_c\gamma_d(\gamma_{cd}^2+\Delta_2^2)+2\gamma_{cd}^2 g_2^2(n+1+n'+1)+g_2^2(n'-n)[g_2^2(n'-n)+i\Delta_2(\gamma_c-\gamma_d)]}\right), \tag{2.6}$$

$$\Delta_1=\omega_1-\nu\,,\quad \Delta_2=\omega_2-\nu\,, \tag{2.7}$$

where ν is the laser frequency and ω_1 and ω_2 are the frequencies of the atomic transitions $a\rightarrow b$ and $c\rightarrow d$. Also,

$$\gamma_{ab}=\tfrac{1}{2}(\gamma_a+\gamma_b),\quad \gamma_{cd}=\tfrac{1}{2}(\gamma_c+\gamma_d)\,. \tag{2.8}$$

The terms with the constant C describe the linear cavity losses,

$$C=\frac{r_1}{\gamma_1}\frac{g^2}{\gamma_{12}}=\frac{\nu}{Q} \tag{2.9}$$

where r_1 and r_2 are the rates of introduction of damping atoms in levels **1** and 2, g is the coupling constant, and $\gamma_{12}=\frac{1}{2}(\gamma_1+\gamma_2)$, where γ_1 and γ_2 are the decay rates for levels 1 and 2 of the atoms. Q is the cavity Q factor. Finally, $n_2=r_2/\gamma_2$ and $n_1=r_1/\gamma_1$.

Specializing to the diagonal elements, we obtain

$$\dot{\rho}_{nn}=T_{\rm I}+T_{\rm II}+T_{\rm III}+T_{\rm IV}+T_{\rm V}+T_{\rm VI}\,, \tag{2.10}$$

where

$$T_{\rm I}=\frac{-A(n+1)}{1+(B/A)(n+1)}\left(\rho_{nn}(t)-\frac{n_b}{n_a}\rho_{n+1,n+1}(t)\right), \tag{2.11}$$

$$T_{\rm II}=\frac{An}{1+(B/A)n}\left(\rho_{n-1,n-1}(t)-\frac{n_b}{n_a}\rho_{nn}(t)\right), \tag{2.12}$$

$$T_{\rm III}=\frac{-G(n+1)}{1+(H/G)(n+1)}\left(\rho_{nn}(t)-\frac{n_d}{n_c}\rho_{n+1,n+1}(t)\right), \tag{2.13}$$

$$T_{\rm IV}=\frac{Gn}{1+(H/G)n}\left(\rho_{n-1,n-1}(t)-\frac{n_d}{n_c}\rho_{nn}(t)\right), \tag{2.14}$$

$$T_{\rm V}=C(n+1)\left(\rho_{n+1,n+1}(t)-\frac{n_2}{n_1}\rho_{nn}(t)\right), \tag{2.15}$$

$$T_{\rm VI}=-Cn\left(\rho_{nn}(t)-\frac{n_2}{n_1}\rho_{n-1\,n-1}(t)\right). \tag{2.16}$$

Here,

$$n_k=r_k/\gamma_k,\quad k=a,b,c,d\,, \tag{2.17}$$

$$\begin{aligned}
A&=2n_a g_1^2[\gamma_{ab}/(\gamma_{ab}^2+\Delta_1^2)]\,,\\
B&=8n_a g_1^4[\gamma_{ab}^3/\gamma_a\gamma_b(\gamma_{ab}^2+\Delta_1^2)^2]\,,\\
G&=2n_c g_2^2[\gamma_{cd}/(\gamma_{cd}^2+\Delta_2^2)]\,,\\
H&=8n_c g_2^4[\gamma_{cd}^3/\gamma_c\gamma_d(\gamma_{cd}^2+\Delta_2^2)^2]\,.
\end{aligned} \tag{2.18}$$

All the terms in Eq. (2.10) have simple physical interpretations. $T_{\rm I}$ has two parts; the first is a loss for the n photon state of the laser mode due to spontaneous and stimulated emission and the second is a gain due to absorption from an $n+1$ photon state. Both contributions are due to atoms of the active medium, but the first is due to atoms introduced in the upper lasing level, while the second is due to atoms introduced in the lower lasing level. $T_{\rm II}$ is also due to the active atoms. $T_{\rm III}$ is from the absorber atoms. The first part describes absorption out of the n photon state of the laser mode due to absorber atoms introduced in the upper levels, while the next part shows an enhancement of the n photon state probability due to those introduced to level d. $T_{\rm IV}$ is similarly interpreted. For the active medium to show gain, $n_a>n_b$, and for the absorber to be an absorber, $n_d>n_c$. $T_{\rm V}$ and $T_{\rm VI}$ represent linear cavity losses.

The fact that we have considered only single photon processes and single atom interactions with the electromagnetic field leads to the detailed

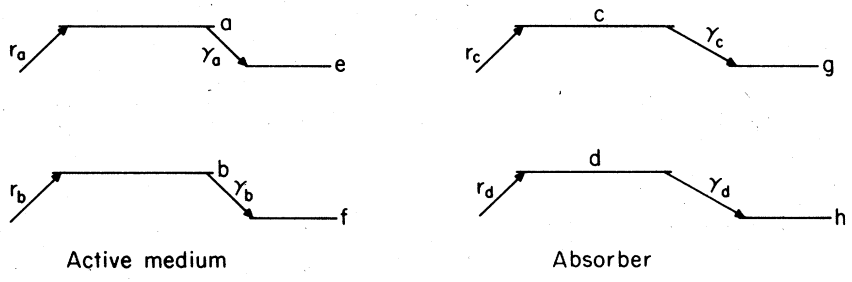

FIG. 2. Energy levels and decays of the active medium and the saturable absorber.

balance condition in the steady state. Thus, when $\dot{\rho}_{nn}=0$, the flow of probability from the n to the $n-1$ photon state equals that from the $n-1$ to the n photon state. A similar condition holds for the n and $n+1$ photon states. The three-term recursion relation simplifies to two identical two-term relations. This relation is

$$\rho_{nn}\left(C+\frac{A}{1+(B/A)n}\frac{n_b}{n_a}+\frac{G}{1+(H/G)n}\frac{n_d}{n_c}\right)=\rho_{n-1,n-1}\left(C\frac{n_2}{n_1}+\frac{A}{1+(B/A)n}+\frac{G}{1+(H/G)n}\right). \quad (2.19)$$

$P(n)(\equiv\rho_{nn})$, the probability distribution for n photons in the laser cavity, now takes the form:

$$P(n)=\mathfrak{N}\prod_{k=0}^{n}\left(\frac{n_2}{n_1}+\frac{A/C}{1+(B/A)k}+\frac{G/C}{1+(H/G)k}\right)\Big/\left(1+\frac{A/C}{1+(B/A)k}\frac{n_b}{n_a}+\frac{G/C}{1+(H/G)k}\frac{n_d}{n_c}\right), \quad (2.20)$$

where $\mathfrak{N}$ is a normalization constant.

Let us summarize the conditions under which this result holds. The model considers an active medium and an absorber which are both homogeneously broadened, stationary two-level atoms. Atoms may be introduced into both upper and lower levels of each type of atom, the lifetimes of which are known. The possible detuning of the laser mode from the atomic transitions has been retained. Linear cavity losses have been considered. We have finally calculated the diagonal elements of the radiation density matrix in the steady state. Coefficients A and B are the gain and saturation coefficients (of the usual Scully-Lamb theory) for the active atoms; G and H are similar terms for the absorber.

We now show the correspondence of (2.2) to the steady-state distribution found by Lugiato *et al.*[10(a)] An approximate form for $P(n)$ can be found when n is large by converting Eq. (2.20) into an approximate differential equation. We have

$$P(n)=P(n)-P(n-1)+P(n-1)=P(n-1)L(n),$$

where $L(n)$ is the quantity after the product sign in Eq. (2.20). For large n

$$\frac{dP(n)}{dn}=P(n-1)[L(n)-1]. \quad (2.21)$$

Therefore,

$$P(n)=\mathfrak{N}_1\exp\left[\int\left(1-\frac{1}{L(n)}\right)dn\right]. \quad (2.22)$$

Using the given form for $L(n)$, we obtain

$$\left[\left(1-\frac{A/C}{1+(B/A)n}(1-n_b/n_a)-\frac{G/C}{1+(H/G)n}(1-n_d/n_c)\right)+\left(\frac{A/C}{1+(B/A)n}+\frac{G/C}{1+(H/G)n}\right)\frac{d}{dn}\right]P(n)=0.$$

If we let $fr^2=n$, where r is the magnitude of the complex amplitude of Lugiato *et al.*, and f a constant quantity, then on neglecting the r dependence of the coefficient of the derivative term (see comment by Lugiato *et al.* in Ref. 10) we get, (i.e., assuming a zero-temperature field reservior in conformity with Ref. 10)

$$\left[Cr\left(1-\frac{A/C}{1+Br^2f/A}(1-n_b/n_a)-\frac{G/C}{1+r^2fH/G}(1-n_d/n_c)\right)+\frac{A+G}{2f}\frac{d}{dr}\right]P(r)=0. \quad (2.23)$$

This equation is of the same form as Eq. (4.12) of Ref. 10(a), from which they find $P(r)$, if we identify $(A+G)/2f$ with q. Note that Eq. (2.23) is derived from (2.20) after several approximations. Equation (2.20) is the more exact expression, which is valid for all n, and which also gives us the r-dependent coefficient for the derivative term in Eq. (2.23). For all practical purposes and for analytic considerations, Eq. (2.23) and its solution are very useful. A more complete analysis of the photon statistics has been given by Lugiato *et al.* in Refs. 10; we have only pointed out an alternative method for obtaining some of their results.

To be able to see clearly the behavior of the LSA, let us consider a simplified case, where the active atoms are introduced in state a only, and the absorber atoms in state d only. The damping atoms are injected only in state 1. Then we get, instead of Eq. (2.10),

$$\dot{\rho}_{nn}=\frac{-A(n+1)}{1+(B/A)(n+1)}\rho_{nn}(t)+\frac{An}{1+(B/A)n}\rho_{n-1,n-1}(t)+\frac{G'(n+1)}{1+(H'/G')(n+1)}\rho_{n+1,n+1}(t)-\frac{G'n}{1+(H'/G')n}\rho_{nn}(t)-Cn\rho_{nn}(t)+C(n+1)\rho_{n+1,n+1}(t), \quad (2.24)$$

where

$$G'=2n_dg_2^2[\gamma_{cd}/(\gamma_{cd}^2+\Delta_2^2)],$$
$$H'=8n_dg_2^4[\gamma_{cd}^3/\gamma_c\gamma_d(\gamma_{cd}^2+\Delta_2^2)^2]. \quad (2.25)$$

Detailed balance now gives us

$$\rho_{nn}=\rho_{n-1,n-1}\frac{A/C}{[1+(B/A)n][1+(G'/C)/(1+H'n/G')]}, \quad (2.26)$$

i.e.,

$$P(n)=\mathfrak{N}_2\prod_{k=0}^{n}\frac{A/C}{(1+kB/A)[1+(G'/C)/(1+kH'/G')]}\cdot \tag{2.27}$$

Using Eq. (2.22),

$$P(n)=\mathfrak{N}_1'\exp\left[n\left(1-\frac{C}{A}-\frac{G'^2B}{A^2H'}\right)-n^2\frac{BC}{2A^2}+\left(\frac{G'^3B}{A^2H'^2}-\frac{G'^2}{AH'}\right)\ln\left(1+\frac{nH'}{G'}\right)\right]. \tag{2.28}$$

Recalling that the fluctuations in the magnitude E of the complex scalar field amplitude are given by $P(E)=\mathfrak{N}e^{-F(E)/R_L\sigma}$ where $\mathfrak{N}$ is a normalization constant, $F(E)$ is the Gibbs potential for the laser field, R_L is a constant characterizing spontaneous emission by the atoms, and σ is the zero-field inversion, the Gibbs potential for a LSA is seen to be

$$F(E)\approx\frac{-(A-C)E^2}{2}+\frac{BCE^4}{4A}+\frac{G'^2}{2H'}\ln\left(1+\frac{E^2H'}{G'}\right), \tag{2.29}$$

where we have taken $E^2\sim n$ and the terms nG'^2B/A^2H' and

$$(G'^3B/A^2H'^2)\ln(1+nH'/G')$$

have been neglected since they are smaller than the others by a factor B/A. If we now let

$$A\equiv a\sigma,\quad B\equiv b\sigma,\quad C\equiv a\sigma_T,\quad G'\equiv a\sigma_s,\quad G'/H'\equiv I_s, \tag{2.30}$$

then, since $C/A\sim 1$,

$$F(E)\approx -\tfrac{1}{2}a[(\sigma-\sigma_T)E^2-\sigma_sI_s\ln(1+E^2/I_s)]+\tfrac{1}{4}b\sigma E^4. \tag{2.31}$$

This is identical to the result of Scott *et al.* in Ref. 5. We now examine the dye-laser photon distribution.

III. DYE LASER

Quite apart from its extreme usefulness in present day physics and chemistry, the dye laser is an interesting system to study in its own right. The energy-level structure of dye molecules with singlet electronic states and the corresponding triplets make the dye laser behave as if it had an absorbing system built into each molecule. The various processes which occur in a dye molecule in solution have been discussed in detail in many texts and papers, e.g., Refs. 8 and 12–14.

We adopt the dye-laser model with energy-level scheme shown in Fig. 3; a brief sketch is now given of the various processes in which a dye molecule may participate during lasing. A molecule introduced in the upper lasing level a may go to the lower lasing level b by stimulated or spontaneous emission into the laser mode of the electromagnetic field. It may also decay radiatively not into the laser mode, or nonradiatively to some other sublevel m of the ground singlet state. Alternatively, the molecule may go over to the lowest triplet state T_1 by an intersystem crossing process. Once in T_1 the molecule may act as an absorber for the laser mode of the radiation field, in which case it goes to the upper triplet state T_2, or it may decay to the ground singlet state by what is called a triplet quenching process. The various processes and rates are marked in Fig. 3. If the molecule reaches T_2 by absorption of the laser radiation, it may be transferred to some other higher triplet or singlet level, or it may drop back into the lower triplet state. In the former case, we say that the molecule is lost to the lasing process, and denote this by the rate γ_f of decay to a fictitious level l. In the Schaefer-Willis model, $\beta\nu_{Tu}$ describes the rate of decay of molecules from the upper triplet to the lower triplet state due to nonradiative processes or radiative processes not concerning the laser radiation mode. The model presented here neglects this process, and we assume the entire loss of molecules from the lasing process to be due to the decay rate $\gamma_f(\equiv\nu_{Tu})$. That is, if in the Schaefer-Willis model one assumes that the entire rate ν_{Tu} describes molecules being lost to the lasing process and that molecules transfer from level T_2 to level T_1 only via radiative processes involving the laser mode,

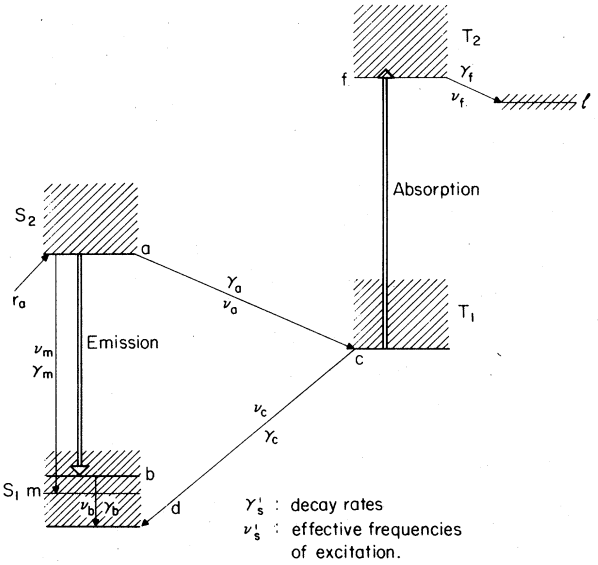

FIG. 3. Energy levels of a dye molecule.

then one puts $\beta = 0$ in their model. The Schaefer-Willis treatment of dye-laser action is based on a laser theory developed by Willis in Ref. 8(c), while our method of analysis follows that of Scully and Lamb.[1(a)]

In the analysis to be given we will assume single-mode operation, and that molecules are introduced only in the upper lasing level at a rate r_a by an unspecified pumping process. The decays γ_a, γ_b, γ_c, γ_m, and γ_f are introduced through interaction of the molecule with fictitious reservoirs. Since these decays do not produce any photons in the laser mode, we trace over the reservoir variables to obtain the relevant laser-radiation density-matrix equation of motion. The fictitious frequencies ν_a, ν_b, ν_c, ν_f, and ν_m (where the indices run over large numbers of modes) belong to the decay-inducing reservoirs.

A. Hamiltonian

The dye-laser Hamiltonian is $\mathcal{H} = \hbar H$, with H given by

$$H = \nu a^\dagger a + \sum_a \nu_a a_a^\dagger a_a + \sum_b \nu_b a_b^\dagger a_b + \sum_c \nu_c a_c^\dagger a_c + \sum_f \nu_f a_f^\dagger a_f + \sum_m \nu_m a_m^\dagger a_m + \sum_{\substack{K=a,b,c,d \\ f,l,m}} \epsilon_K A_K^\dagger A_K + g_1(a^\dagger A_b^\dagger A_a + a A_a^\dagger A_b)$$

$$+ g_2(a^\dagger A_c^\dagger A_f + a A_f^\dagger A_c) + \sum_a g_a(a_a^\dagger A_c^\dagger A_a + a_a A_a^\dagger A_c) + \sum_b g_b(a_b^\dagger A_d^\dagger A_b + a_b A_b^\dagger A_d) + \sum_c g_c(a_c^\dagger A_d^\dagger A_c + a_c A_c^\dagger A_d)$$

$$+ \sum_f g_f(a_f^\dagger A_l^\dagger A_f + a_f A_f^\dagger A_l) + \sum_m g_m(a_m^\dagger A_m^\dagger A_a + a_m A_a^\dagger A_m) \,, \tag{3.1}$$

i.e.,

$$H = H_0 + H_0^a + H_0^b + H_0^c + H_0^f + H_0^m + V_1 + V_2 + \sum_a V^a + \sum_b V^b + \sum_c V^c + \sum_f V^f + \sum_m V^m \,.$$

The free laser field and molecule Hamiltonians are

$$\hbar H_0 = \hbar \nu a^\dagger a + \hbar \sum_{\substack{K=a,b,c,d \\ f,l,m}} \epsilon_K A_K^\dagger A_K \,.$$

The creation and annihilation operators for the laser radiation are $a^\dagger$ and a. The $A_K^\dagger$'s and A_K's are the corresponding operators for the molecular levels. $\hbar H_0^a \equiv \hbar \sum_a \nu_a a_a^\dagger a_a$ etc. are the energies of the reservoir excitations.

$$V_{1\,(2)} \equiv g_{1\,(2)}(a^\dagger A_{b\,(c)}^\dagger A_{a\,(f)} + a A_{a\,(f)}^\dagger A_{b\,(c)})$$

describes the interaction of the laser radiation with the upper and lower lasing levels and the absorbing triplet levels.

$$g_{1\,(2)} = \frac{ex_{ab(cf)}\mathcal{E}}{\sqrt{2}\,\hbar} \text{ and } \mathcal{E} = \left(\frac{\hbar\Omega}{LA\epsilon_0}\right)^{1/2} ,$$

where ex_{ab}, ex_{cf} are the dipole-moment matrix elements.

$$\sum_a V^a \equiv \sum_a g_a(a_a^\dagger A_c^\dagger A_a + a_a A_a^\dagger A_c) \,, \quad \text{etc.} \,,$$

describe the interaction of the molecule with the reservoirs, which introduce (in the Wigner-Weisskopf approximation) phenomenological decay constants into the problem.

The lasing transition and the triplet absorbing transition frequencies are assumed to be the same.

B. Equations of motion for the density-matrix elements

The procedure followed here is that of Scully and Lamb. The total system density-matrix equation of motion in the interaction picture is

$$\dot{\rho}_T(t) = -i[V_1(t), \rho_T] - i[V_2(t), \rho_T(t)]$$

$$- i\left[\sum_a V^a + \sum_b V^b + \sum_c V^c + \sum_f V^f + \sum_m V^m, \rho_T\right]. \tag{3.2a}$$

The equations for the matrix elements relevant to our problem are obtained after several approximation procedures, which are given in the Appendix. After tracing over the unobserved or reservoir variables we obtain the following set of equations in the Schrödinger picture.

$$\dot{\rho}_{an;an'} = -i[H_0 + V_1, \rho]_{an;an'} - \gamma_a \rho_{an;an'} - \gamma_m \rho_{an;an'} \,,$$

$$\dot{\rho}_{bn+1;bn'+1} = -i[H_0 + V_1, \rho]_{bn+1;bn'+1} - \gamma_b \rho_{bn+1;bn'+1} \,,$$

$$\dot{\rho}_{bn+1;an'} = -i[H_0 + V_1, \rho]_{bn+1;an'} - \gamma_{ab} \rho_{bn+1;an'} \,,$$

$$\dot{\rho}_{cn;cn'} = \gamma_a \rho_{an;an'} - \gamma_c \rho_{cn;cn'} - i[H_0 + V_2, \rho]_{cn,cn'} \,,$$

$$\dot{\rho}_{fn-1;fn'-1} = -\gamma_f \rho_{fn-1;fn'-1} - i[H_0 + V_2, \rho]_{fn-1;fn'-1} \,,$$

$$\dot{\rho}_{fn-1;\,cn'} = -\gamma_{cf}\rho_{fn-1;\,cn'} - i[H_0 + V_2, \rho]\,,$$

$$\dot{\rho}_{ln-1;\,ln'-1} = \gamma_f \rho_{fn-1;\,fn'-1}\,,$$

$$\dot{\rho}_{dn+1;\,dn'+1} = \gamma_b \rho_{bn+1;\,bn'+1}$$

$$\dot{\rho}_{dn;\,dn'} = \gamma_c \rho_{cn;\,cn'}$$

i.e.,

$$\dot{\rho}_{dn;\,dn'} = \gamma_b \rho_{bn;\,bn'} + \gamma_c \rho_{cn;\,cn'}\,,$$

$$\dot{\rho}_{mn;\,mn'} = \gamma_m \rho_{an;\,an'}\,.$$

$$\gamma_{ab} = \tfrac{1}{2}(\gamma_a + \gamma_b + \gamma_m), \quad \gamma_{cf} = \tfrac{1}{2}(\gamma_c + \gamma_f + \gamma_a) \qquad (3.2b)$$

and γ_a, γ_b, γ_m, γ_c, and γ_f are defined explicitly in the Appendix. The equations for levels l, d, and m can be integrated from t_0, the time of introduction of the molecule to $t_0 + T$, where T is larger than all the decay times involved, but small compared to the times for growth or decay of the field. Then,

$$\begin{aligned}
\rho_{ln-1;\,ln'-1}(t_0+T) &= \gamma_f \int_{t_0}^{t_0+T} dt'\, \rho_{fn-1;\,fn'-1}(t')\,,\\
\rho_{dn+1;\,dn'+1}(t_0+T) &= \gamma_b \int_{t_0}^{t_0+T} \rho_{bn+1;\,bn'+1}(t')\,dt'\,,\\
\rho_{dn;\,dn'}(t_0+T) &= \gamma_c \int_{t_0}^{t_0+T} \rho_{cn;\,cn'}(t')\,dt'\,,\\
\rho_{mn;\,mn'}(t_0+T) &= \gamma_m \int_{t_0}^{t_0+T} \rho_{an;\,an'}(t')\,dt'\,,
\end{aligned} \qquad (3.3)$$

Expanding out the commutators with H_0 (ν = laser mode frequency; $\omega = \epsilon_a - \epsilon_b = \epsilon_c - \epsilon_d$),

$$\begin{aligned}
\dot{\rho}_{an;\,an'} &= -i[(n-n')\nu - i(\gamma_a+\gamma_m)]\rho_{an;\,an'}\\
&\quad - i(V_{1an;\,bn+1}\rho_{bn+1;\,an'} - \rho_{an;\,bn'+1}V_{1bn'+1;\,an'})\,,\\
\dot{\rho}_{bn+1;\,bn'+1} &= -i[(n-n')\nu - i\gamma_b]\rho_{bn+1;\,bn'+1}\\
&\quad - i(V_{1bn+1;\,an}\rho_{an;\,bn'+1} - \rho_{bn+1;\,an'}V_{1an';\,bn'+1})\,,\\
\dot{\rho}_{bn+1;\,an'} &= -i[(n-n')\nu - (\omega-\nu) - i\gamma_{ab}]\rho_{bn+1;\,an'}\\
&\quad - i(V_{1bn+1;\,an}\rho_{an;\,an'} - \rho_{bn+1;\,bn'+1}V_{1bn'+1;\,an'})\,,\\
\dot{\rho}_{an;\,bn'+1} &= -i[(n-n')\nu + (\omega-\nu) - i\gamma_{ab}]\rho_{an;\,bn'+1}\\
&\quad - i(V_{1an;\,bn+1}\rho_{bn+1;\,bn'+1} - \rho_{an;\,an'}V_{1an';\,bn'+1})\,,\\
\dot{\rho}_{cn;\,cn'} &= -i[(n-n')\nu - i\gamma_c]\rho_{cn;\,cn'} + \gamma_a\rho_{an;\,an'}\\
&\quad - i(V_{2cn;\,fn-1}\rho_{fn-1;\,cn'} - \rho_{cn;\,fn'-1}V_{2fn'-1;\,cn'})\,,\\
\dot{\rho}_{fn-1;\,fn'-1} &= -i[(n-n')\nu - i\gamma_f]\rho_{fn-1;\,fn'-1}\\
&\quad - i(V_{2fn-1;\,cn}\rho_{cn;\,fn'-1} - \rho_{fn-1;\,cn'}V_{2cn';\,fn'-1})\,,\\
\dot{\rho}_{cn;\,fn'-1} &= -i[(n-n')\nu - (\omega-\nu) - i\gamma_{cf}]\rho_{cn;\,fn'-1}\\
&\quad - i(V_{2cn;\,fn-1}\rho_{fn-1;\,fn'-1} - \rho_{cn;\,cn'}V_{2cn';\,fn'-1})\,,\\
\dot{\rho}_{fn-1;\,cn'} &= -i[(n-n')\nu - (\omega-\nu) - i\gamma_{cf}]\rho_{fn-1;\,cn'}\\
&\quad - i(V_{2fn-1;\,cn}\rho_{cn;\,cn'} - \rho_{fn-1;\,fn'-1}V_{2fn'-1;\,cn'})\,.
\end{aligned} \qquad (3.4)$$

Going to the interaction picture for the radiation density matrix only, and replacing $\rho_{nn'}$ by $\rho_{nn'}e^{-i(n-n')t\nu}$ and using the following notation,

$$\begin{aligned}
&\sigma_{11} \equiv \sigma_{an;\,an'}\,, \quad \sigma_{33} \equiv \sigma_{fn-1;\,fn'-1}\,,\\
&\sigma_{12} \equiv \sigma_{an;\,bn'+1}\,, \quad \sigma_{34} \equiv \sigma_{fn-1;\,cn'}\,,\\
&\sigma_{21} \equiv \sigma_{bn+1;\,an'}, \quad \sigma_{43} \equiv \sigma_{cn;\,fn'-1}\,,\\
&\sigma_{22} \equiv \sigma_{bn+1;\,bn'+1}\,, \quad \sigma_{44} \equiv \sigma_{cn;\,cn'}\,,
\end{aligned} \qquad (3.5)$$

where

$$\int_{t_0}^{t_0+T} \rho_{\beta\beta'}(t')\,dt' = \sigma_{\beta\beta'}(t_0+T), \quad \beta = 1,2, \quad \beta' = 1,2$$

and $\int_{t_0}^{t_0+T} \rho_{\alpha\alpha'}(t')\,dt' = \sigma_{\alpha\alpha'}(t_0+T)$,

$\alpha = 3,4$, $\alpha' = 3,4$,

we obtain, on integration from t_0 to t_0+T, the equations:

$$\begin{aligned}
0 - \rho_{an;\,an'}(t_0) &= -(\gamma_a+\gamma_m)\sigma_{11} - i(V_{1an;\,bn+1}\sigma_{21}\\
&\quad - \sigma_{12}V_{1bn'+1;\,an'})\,,\\
0 &= -[i(\omega-\nu) + \gamma_{ab}]\sigma_{12}\\
&\quad - i(V_{1an;\,bn+1}\sigma_{22} - \sigma_{11}V_{1an';\,bn'+1})\,,\\
0 &= -[-i(\omega-\nu) + \gamma_{ab}]\sigma_{21}\\
&\quad - i(V_{1bn+1;\,an}\sigma_{11} - \sigma_{22}V_{1bn'+1;\,an'})\,,\\
0 &= -\gamma_b\sigma_{22} - i(V_{1bn+1;\,an}\sigma_{12} - \sigma_{21}V_{1an';\,bn'+1})\,,\\
0 &= -\gamma_f\sigma_{33} - i(V_{2fn-1;\,cn}\sigma_{43} - \sigma_{34}V_{2cn';\,fn'-1})\,,\\
0 &= -i[-i(\omega-\nu) + \gamma_{cf}]\sigma_{43}\\
&\quad - i(V_{2cn;\,fn-1}\sigma_{33} - \sigma_{44}V_{2cn';\,fn'-1})\,,\\
0 &= -[i(\omega-\nu) + \gamma_{cf}]\sigma_{34}\\
&\quad - i(V_{2fn-1;\,cn}\sigma_{44} - \sigma_{33}V_{2fn'-1;\,cn'})\,,\\
0 &= -\gamma_c\sigma_{44} + \gamma_a\sigma_{11} - i(V_{2cn;\,fn-1}\sigma_{34} - \sigma_{43}V_{2fn'-1;\,cn'})\,.
\end{aligned} \qquad (3.6)$$

The initial condition that the molecule is injected in level a gives us the condition

$$\rho_{nn'}(t_0) = \rho_{an;\,an'}(t_0)\,. \qquad (3.7)$$

Also,

$$\begin{aligned}
\rho_{nn'}(t_0+T) &= \rho_{an;\,an'}(t_0+T) + \rho_{bn;\,bn'}(t_0+T)\\
&\quad + \rho_{cn;\,cn'}(t_0+T) + \rho_{dn;\,dn'}(t_0+T)\\
&\quad + \rho_{fn;\,fn'}(t_0+T) + \rho_{ln;\,ln'}(t_0+T)\\
&\quad + \rho_{mn;\,mn'}(t_0+T)\,.
\end{aligned}$$

At time t_0+T all elements except $\rho_{an;an'}(t_0+T)$, $\rho_{ln;ln'}(t_0+T)$, and $\rho_{mn;mn'}(t_0+T)$ are negligibly small, since T is taken to be larger than all molecular decay times involved (and smaller than any decay or growth time of the laser mode).

Using Eq. (3.3) and Eq. (3.5), we obtain

$$\rho_{nn'}(t_0+T) = \underset{\substack{(n\to n+1)\\(n'\to n'+1)}}{\gamma_f\sigma_{33}} + \underset{\substack{(n\to n-1)\\(n'\to n'-1)}}{\gamma_b\sigma_{22}} + \gamma_c\sigma_{44} + \gamma_m\sigma_{11} \tag{3.8}$$

In matrix form, Eqs. (3.6) are [with $\Delta \equiv (\omega - \nu)$]

$$\begin{pmatrix}
-i(\gamma_a+\gamma_m) & -V_{1\,bn'+1;\,an'} & V_{1an;\,bn+1} & 0 & 0 & 0 & 0 & 0 \\
-V_{1an';\,bn'+1} & -i\gamma_{ab}+\Delta & 0 & V_{1an;\,bn+1} & 0 & 0 & 0 & 0 \\
V_{1bn+1;\,an} & 0 & -\Delta-i\gamma_{ab} & -V_{1bn'+1;\,an'} & 0 & 0 & 0 & 0 \\
0 & V_{1bn+1;\,an} & -V_{1an';\,bn'+1} & -i\gamma_b & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -i\gamma_f & -V_{2cn';\,fn'-1} & V_{2fn-1;\,cn} & 0 \\
0 & 0 & 0 & 0 & -V_{2fn'-1;\,cn'} & \Delta-i\gamma_{cf} & 0 & V_{2fn-1;\,cn} \\
0 & 0 & 0 & 0 & V_{2cn;\,fn-1} & 0 & -\Delta-i\gamma_{cf} & -V_{2cn';\,fn'-1} \\
i\gamma_a & 0 & 0 & 0 & 0 & V_{2cn;\,fn-1} & -V_{2fn'-1;\,cn'} & -i\gamma_c
\end{pmatrix}
\begin{pmatrix} \sigma_{11} \\ \sigma_{12} \\ \sigma_{21} \\ \sigma_{22} \\ \sigma_{33} \\ \sigma_{34} \\ \sigma_{43} \\ \sigma_{44} \end{pmatrix}$$

$$= \begin{pmatrix} -i\rho_{nn'}(t_0) \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \\ 0 \end{pmatrix} . \tag{3.9}$$

The first four equations are not coupled to the last four; they form a closed set (though the solution from these for σ_{11} is required for the last four). The solutions are those of Scully and Lamb:

$$(\gamma_a+\gamma_m)\sigma_{11} = \rho_{nn'}(t_0) - [(n+1)\mathcal{R}_{nn'} + (n'+1)\mathcal{R}^*_{n'n}]\rho_{nn'}(t_0) , \tag{3.10}$$

where

$$\mathcal{R}_{nn'} = \frac{g_1^2\gamma_b(\gamma_{ab}+i\Delta)+g_1^4(n'-n)}{(\gamma_a+\gamma_m)\gamma_b(\gamma_{ab}^2+\Delta^2)+g_1^2\gamma_{ab}(\gamma_a+\gamma_b+\gamma_m)(n+1+n'+1)+g_1^2(n-n')[g_1^2(n-n')+i\Delta(\gamma_a+\gamma_b+\gamma_m)]} \tag{3.11}$$

and

$$\gamma_b\sigma_{22} = (\mathcal{R}_{nn'}+\mathcal{R}^*_{n'n})[(n+1)(n'+1)]^{1/2}\rho_{nn'}(t_0) . \tag{3.12}$$

The solution simplifies considerably when we wish to look at the diagonal, on-resonance case; then,

$$\gamma_m\sigma_{11} = \frac{\gamma_m}{\gamma_a+\gamma_m}\rho_{nn}\left(1 - \frac{2(n+1)\gamma_b\gamma_{ab}g_1^2}{(\gamma_a+\gamma_m)\gamma_b\gamma_{ab}^2+2g_1^2(n+1)(\gamma_a+\gamma_b+\gamma_m)\gamma_{ab}}\right) \tag{3.13}$$

and

$$\underset{\substack{(n\to n-1)\\(n'\to n'-1)}}{\gamma_b\sigma_{22}} = \frac{2n\gamma_b\gamma_{ab}g_1^2\rho_{n-1,n-1}}{(\gamma_a+\gamma_m)\gamma_b\gamma_{ab}^2+2g_1^2n\gamma_{ab}(\gamma_a+\gamma_b+\gamma_m)} . \tag{3.14}$$

The solution of the triplet equations is

$$\gamma_f\sigma_{33}=\frac{2i\gamma_f\gamma_{cf}g_2^2(nn')^{1/2}(-i\gamma_a\sigma_{11})}{\gamma_c\gamma_f(\gamma_{cf}^2+\Delta^2)+g_2^2(n'+n)\gamma_{cf}(\gamma_c+\gamma_f)+g_2^2(n'-n)[g_2^2(n'-n)+i\Delta(\gamma_f-\gamma_c)]} \tag{3.15}$$

$$\gamma_c\sigma_{44}=\frac{\gamma_c(-i\gamma_a\sigma_{11})[i\gamma_f(\Delta^2+\gamma_{cf}^2)+g_2^2n'(\Delta+i\gamma_{cf})-g_2^2n(\Delta-i\gamma_{cf})]}{\gamma_c\gamma_f(\gamma_{cf}^2+\Delta^2)+g_2^2\gamma_{cf}(n'+n)(\gamma_c+\gamma_f)+g_2^2(n'-n)[g_2^2(n'-n)+i\Delta(\gamma_f-\gamma_c)]} \tag{3.16}$$

which for the on-diagonal, on-resonance matrix elements gives

$$\gamma_f\sigma_{\substack{33\\(n\to n+1\\n'\to n'+1)}}=\frac{2\gamma_f\gamma_{cf}g_2^2(n+1)\rho_{n+1,n+1}}{\gamma_c\gamma_f\gamma_{cf}^2+g_2^2\gamma_{cf}(\gamma_c+\gamma_f)2(n+1)}\frac{\gamma_a}{\gamma_a+\gamma_m}\left(1-\frac{2(n+2)\gamma_b\gamma_{ab}g_1^2}{(\gamma_a+\gamma_m)\gamma_b\gamma_{ab}^2+2(n+2)g_1^2(\gamma_a+\gamma_b+\gamma_m)\gamma_{ab}}\right), \tag{3.17}$$

$$\gamma_c\sigma_{44}=\rho_{nn}\frac{\gamma_c\gamma_f\gamma_{cf}^2+2g_2^2\gamma_c\gamma_{cf}n}{\gamma_c\gamma_f\gamma_{cf}^2+2ng_2^2\gamma_{cf}(\gamma_c+\gamma_f)}\frac{\gamma_a}{\gamma_a+\gamma_m}\left(1-\frac{2(n+1)\gamma_b\gamma_{ab}g_1^2}{(\gamma_a+\gamma_m)\gamma_b\gamma_{ab}^2+2g_1^2(n+1)(\gamma_a+\gamma_b+\gamma_m)\gamma_{ab}}\right). \tag{3.18}$$

Using the coarse-graining approximation,[1,4] we obtain

$$\dot\rho_{nn}=r_a[\rho_{nn}(t_0+T)-\rho_{nn}(t_0)] \tag{3.19}$$

If we include the loss terms for the cavity and use Eqs. (3.8) and (3.19), then with the notation

$$\begin{aligned}
&A\equiv 2g_1^2r_a/(\gamma_a+\gamma_m)\gamma_{ab}\,,\\
&B\equiv 4g_1^4r_a(\gamma_a+\gamma_b+\gamma_m)/(\gamma_a+\gamma_m)^2\gamma_b\gamma_{ab}^2\,,\\
&G\equiv 2g_2^2r_a/\gamma_c\gamma_{cf}\,,\\
&H\equiv 4g_2^4r_a(\gamma_c+\gamma_f)/\gamma_f\gamma_c^2\gamma_{cf}^2\,,
\end{aligned} \tag{3.20}$$

we obtain on performing some algebra,

$$\begin{aligned}
\dot\rho_{nn}=&\rho_{n-1,n-1}\frac{An}{1+(B/A)n}+\rho_{nn}r_a\left(1-\frac{\gamma_m}{\gamma_a+\gamma_m}\right)\left(1-\frac{A(n+1)/r_a}{1+(B/A)(n+1)}\right)\\
&-\rho_{nn}r_a-\rho_{nn}\frac{Gn}{1+(H/G)n}\left(\frac{\gamma_a}{\gamma_a+\gamma_m}\right)\left(1-\frac{A(n+1)/r_a}{1+(B/A)(n+1)}\right)\\
&+\rho_{nn}r_a\left(\frac{\gamma_m}{\gamma_a+\gamma_m}\right)\left(1-\frac{A(n+1)/r_a}{1+(B/A)(n+1)}\right)+\rho_{n+1,n+1}\frac{G(n+1)}{1+(H/G)(n+1)}\left(\frac{\gamma_a}{\gamma_a+\gamma_m}\right)\left(1-\frac{A(n+2)/r_a}{1+(B/A)(n+2)}\right)\\
&-Cn\rho_{nn}+C(n+1)\rho_{n+1,n+1}\,.
\end{aligned}$$

After some more algebra, we obtain

$$\begin{aligned}
\dot\rho_{nn}=&\rho_{n-1,n-1}\frac{An}{1+(B/A)n}-\rho_{nn}\frac{A(n+1)}{1+(B/A)(n+1)}-\rho_{nn}\frac{Gn}{1+(H/G)n}\left(1-\frac{A(n+1)/r_a}{1+(B/A)(n+1)}\right)\left(\frac{\gamma_a}{\gamma_a+\gamma_m}\right)\\
&+\rho_{n+1,n+1}\frac{G(n+1)}{1+(H/G)(n+1)}\left(1-\frac{A(n+2)/r_a}{1+(B/A)(n+2)}\right)\left(\frac{\gamma_a}{\gamma_a+\gamma_m}\right)-Cn\rho_{nn}+C(n+1)\rho_{n+1,n+1}
\end{aligned} \tag{3.21}$$

which can be written as

$$\dot\rho_{nn}=T_{\rm I}+T_{\rm II}+T_{\rm III}+T_{\rm IV}+T_{\rm V}+T_{\rm VI}\,.$$

In the steady state, detailed balance prevails, giving a one-step recursion relation:

$$An\left(\frac{1}{1+(B/A)n}\right)\rho_{n-1,n-1}=\rho_{nn}\left[Cn+\frac{Gn}{1+(H/G)n}\left(1-\frac{A(n+1)/r_a}{1+(B/A)(n+1)}\right)\left(\frac{\gamma_a}{\gamma_a+\gamma_m}\right)\right],$$

i.e., with $P(n)\equiv\rho_{nn}$,

$$P(n)=P(n-1)\frac{A}{C}\Big/\left[1+(B/A)n\left[1+\frac{G/C}{1+(H/G)n}\left(\frac{\gamma_a}{\gamma_a+\gamma_m}\right)\left\{1-\frac{A(n+1)/r_a}{1+(B/A)(n+1)}\right\}\right]\right]. \tag{3.22}$$

This is the recursion relation which gives us the dye-laser photon statistics.

Equation (3.21) has a very direct physical interpretation. The first term represents the flow of probability into the n photon state from the $n-1$ photon state, by stimulated and spontaneous emission. This flow is given by the rate of injection of atoms into the upper lasing level times the probability of emission by $n-1$ photons times the probability of $n-1$ photons. The second term represents the flow of probability away from the n photon state due to emission. The third and fourth terms do not occur in the usual laser theory. They represent the flow of probability out of and into the n photon state due to absorption in the triplet states. The third term can be interpreted as being the rate of injection of atoms into state a times the probability of not going to level b through emission times the probability of not decaying to some nonlasing level in S_1 times the probability of absorption in the triplets times the probability of having n photons. The fifth and sixth terms have the usual interpretation for decay terms.[4] Each term thus has a simple interpretation, which allows us to see clearly the processes going on in the dye laser.

If the term in the braces in the denominator of the right-hand side of Eq. (3.22) is replaced by 1, then we obtain the same photon statistics for the dye laser as we did for the laser with a saturable absorber in the approximate expression Eq. (2.27). The similarity between these kinds of lasers shows up clearly in the photon-number distributions.

The correspondence of these results to the results of Schaefer and Willis are apparent if one puts $\gamma_a = k_{ST}$, $\gamma_b = \nu_{Sl}$, $\gamma_m = \nu_{Su}$, $\gamma_c = \nu_{Tl}$, $\gamma_f = \nu_{Tu}$, and $\beta = 0$. This is the notation usually used in dye-laser work. Then, letting

$$A = \frac{SN\nu_e}{\nu_{Su} + k_{ST}}, \quad B = \frac{S^2(\nu_{Sl} + \nu_{SU} + k_{ST})N\nu_e}{(\nu_{SU} + k_{ST})^2 \nu_{Sl}}$$

$$G = \frac{T}{\nu_{Tl}} N\nu_e, \quad H = \frac{T^2(\nu_{Tl} + \nu_{Tu})N\nu_e}{\nu_{Tl}^2 \nu_{Tu}},$$

where these terms are all defined in the first of Ref. 8, we obtain a form for $P(n)$ identical with their form for $R(n)$, the photon-number distribution. We refer the reader to Refs. 8 and 9 for detailed discussions of hysteresis and bistability in dye lasers. Graphs of the second factorial moment of the photon number vs the pumping parameter were presented in Ref. 15.

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APPENDIX: DERIVATION OF EQS. (3.2) FROM THE HAMILTONIAN (3.1)

The states we wish to consider are (for n', $n'+1$, and $n'-1$ we put primes on the Greek letters):

$$\begin{aligned}
&(a, n, 0_a, 0_m) = \alpha, \\
&(b, n+1, 0_b) = \beta, \\
&(m, n, 1_m) = \tau, \\
&(c, n, 1_a, 0_c) = \gamma, \\
&(d, n+1, 1_b) = \delta_b, \\
&(f, n-1, 1_a, 0_f = \phi, \\
&(l, n-1, 1_a, 1_f) = \lambda, \\
&(d, n, 1_a, 1_c) = \delta_c,
\end{aligned} \tag{A1}$$

where the quantities refer to molecular, field, and reservoir states, respectively. Then, using Eq. (3.2a), we obtain

$$\begin{aligned}
\dot{\rho}_{\alpha\alpha'} &= -i[V_1, \rho]_{\alpha\alpha'} - i\sum_s (V^a_{\alpha\gamma}\rho_{\gamma\alpha'} - \rho_{\alpha\gamma'}V^a_{\gamma'\alpha'}) \\
&\quad - i\sum_m (V^m_{\alpha\tau}\rho_{\tau\alpha'} - \rho_{\alpha\tau'}V^m_{\tau'\alpha'}), \\
\dot{\rho}_{\beta\beta'} &= -i[V_1, \rho]_{\beta\beta'} - i\sum_b (V^b_{\beta\delta_b}\rho_{\delta_b\beta'} - \rho_{\beta\delta'_b}V^b_{\delta'_b\beta'}), \\
\dot{\rho}_{\beta\alpha'} &= -i[V_1, \rho]_{\beta\alpha'} - i\Big(\sum_b V^b_{\beta\delta_b}\rho_{\delta_b\alpha'} \\
&\quad - \sum_a \rho_{\beta\gamma'}V^a_{\gamma'\alpha'} - \sum_m \rho_{\beta\tau'}V^m_{\tau'\alpha'}\Big), \\
\dot{\rho}_{\delta_b\delta'_b} &= -i(V^c_{\delta_b\gamma}\rho_{\gamma\delta_c} - \rho_{\delta_c\gamma'}V^c_{\gamma'\delta'_c}), \\
\dot{\rho}_{\delta_c\delta'_c} &= -i(V^c_{\delta_c\gamma}\rho_{\gamma\delta'_c} - \rho_{\delta_c\gamma'}V^c_{\gamma'\delta'_c}), \\
\dot{\rho}_{\gamma\gamma'} &= -i(V^a_{\gamma\alpha}\rho_{\alpha\gamma'} - \rho_{\gamma\alpha'}V^a_{\alpha'\gamma'}) \\
&\quad - i\sum_c (V^c_{\gamma\delta_c}\rho_{\delta_c\gamma'} - \rho_{\gamma\delta'_c}V^c_{\delta'_c\gamma'}) - [V_2, \rho]_{\gamma\gamma'}, \\
\dot{\rho}_{\phi\phi'} &= -i[V_2\rho]_{\phi\phi'} - i\sum_f (V^f_{\phi\lambda}\rho_{\lambda\phi'} - \rho_{\phi\lambda'}V^f_{\lambda'\phi'}), \\
\dot{\rho}_{\lambda\lambda'} &= -i(V^f_{\lambda\phi}\rho_{\phi\lambda'} - \rho_{\lambda\phi'}V^f_{\phi\lambda'}), \\
\dot{\rho}_{\gamma\phi'} &= -i[V_2, \rho]_{\gamma\phi'} \\
&\quad - i(V^a_{\gamma\alpha}\rho_{\alpha\phi'} + \sum_c V^c_{\gamma\delta_c}\rho_{\delta_c\phi'} - \sum_f \rho_{\gamma\lambda'}V^f_{\lambda'\phi'}), \\
\dot{\rho}_{\tau\tau'} &= -i(V^m_{\tau\alpha}\rho_{\alpha\tau'} - \rho_{\tau\alpha'}V^m_{\alpha'\tau'}).
\end{aligned} \tag{A2}$$

Here, ρ is the total system density matrix. Now

we write the following equations; these elements are required in Eqs. (A2):

$$
\begin{aligned}
&\dot{\rho}_{\gamma\alpha'} = -i(V^a_{\gamma\alpha}\rho_{\alpha\alpha'} - \rho_{\gamma\gamma'}V^a_{\gamma'\alpha'}) - iV^c_{\gamma\delta_c}\rho_{\delta_c\alpha'} + i\rho_{\gamma\tau'}V^m_{\tau'\alpha'}\,,\\
&\dot{\rho}_{\delta_b\beta'} = -i(V^b_{\delta_b\beta}\rho_{\beta\beta'} - \rho_{\delta_b\delta'_b}V^b_{\delta'_b\beta'})\,,\\
&\dot{\rho}_{\beta\gamma'} = -i(V^b_{\beta\delta_b}\rho_{\delta_b\gamma'} - \rho_{\beta\alpha'}V^a_{\alpha'\gamma'}) - i(-\rho_{\beta\delta'_c}V^c_{\delta'_c\gamma'})\,,\\
&\dot{\rho}_{\delta_b\alpha'} = -i(V^b_{\delta_b\beta}\rho_{\beta\alpha'} - \rho_{\delta_b\gamma'}V^a_{\gamma'\alpha'}) + i\rho_{\delta_b\tau'}V^m_{\tau'\alpha'}\,,\\
&\dot{\rho}_{\gamma\delta'_c} = -i(V^c_{\gamma\delta_c}\rho_{\delta_c\delta'_c} - \rho_{\gamma\gamma'}V^c_{\gamma'\delta'_c}) - iV^a_{\gamma\alpha}\rho_{\alpha\delta'_c}\,,\\
&\dot{\rho}_{\lambda\phi'} = -i(V^f_{\lambda\phi}\rho_{\phi\phi'} - \rho_{\lambda\lambda'}V^f_{\lambda'\phi'})\,,\\
&\dot{\rho}_{\delta_c\phi'} = -i(V^c_{\delta_c\gamma}\rho_{\gamma\phi'} - \rho_{\delta_c\lambda'}V^f_{\lambda'\phi'})\,,\\
&\dot{\rho}_{\gamma\lambda'} = -i(V^a_{\gamma\alpha}\rho_{\alpha\lambda'} - \rho_{\gamma\phi'}V^f_{\phi'\lambda'}) - i(V^c_{\gamma\delta_c}\rho_{\delta_c\lambda'})\,,\\
&\dot{\rho}_{\alpha\phi'} = -i(V^a_{\alpha\gamma}\rho_{\gamma\phi'} - \rho_{\alpha\lambda'}V^f_{\lambda'\phi'}) - iV^n_{\alpha\tau}\rho_{\tau\phi'}\,,\\
&\dot{\rho}_{\tau\alpha'} = -i(V^m_{\tau\alpha}\rho_{\alpha\alpha'} - \rho_{\tau\tau'}V^m_{\tau'\alpha'})\,.
\end{aligned}
\tag{A3}
$$

These equations are solved by a decoupling approximation, e.g.,

$$\rho_{\alpha\alpha'}(t') = \rho_{an0_a0_m} \simeq \rho_{an;an'}\rho_{0_a0_a}\rho_{0_m0_m}\,.$$

Now

$$\rho_{00}(t') \simeq \rho_{00}(t_0) = 1\,,$$

for both a and m reservoirs. Therefore,

$$\rho_{\alpha\alpha'}(t') = \rho_{an;an'}\,.$$

Similarly,

$$\rho_{\gamma\gamma'} = \rho_{cn1_a;cn'1_a} = \rho_{cn;cn'}\rho_{1_a1_a}\,,$$

$$\rho_{1_a1_a}(t') \simeq \rho_{1_a1_a}(t_0) = 0$$

Therefore $\rho_{\gamma\gamma'}(t) = 0$, in the first of Eqs. (A3). When $\rho_{\gamma\gamma'}$ has to be evaluated in the fifth of these equations, however, we take $\rho_{1_a1_a}(t') = 1$, since the molecule has made a transition to the triplets. Making similar approximations for the other terms, we get

$$
\begin{aligned}
&\rho_{\gamma\alpha'} = -i\int_{t_0}^{t} dt'\, V^a_{\gamma\alpha}\rho_{an;an'}\,,\\
&\rho_{\delta_b\beta'} = -i\int_{t_0}^{t} dt'\, V^b_{\delta_b\beta}\rho_{bn+1;bn'+1}\,,\\
&\rho_{\beta\gamma'} = i\int_{t_0}^{t} dt'\, \rho_{bn+1;an'}V^a_{\alpha'\gamma'}\,,\\
&\rho_{\delta_b\alpha'} = -i\int_{t_0}^{t} dt'\, V^b_{\delta_b\beta}\rho_{bn+1;an'}\,,\\
&\rho_{\gamma\delta'_c} = i\int_{t_0}^{t} dt'\, \rho_{cn;cn'}V^c_{\gamma'\delta'_c}\,,\\
&\rho_{\lambda\phi'} = -i\int_{t_0}^{t} dt'\, V^f_{\lambda\phi}\rho_{fn-1;fn'-1}\,,\\
&\rho_{\delta_c\phi'} = -i\int_{t_0}^{t} dt'\, V^c_{\delta_c\gamma}\rho_{cn;fn'-1}\,,\\
&\rho_{\gamma\lambda'} = i\int_{t_0}^{t} dt'\, V^a_{\alpha\gamma}\rho_{cn;fn'-1}\,,\\
&\rho_{\tau\alpha'} = -i\int_{t_0}^{t} dt'\, V^m_{\tau\alpha}\rho_{an;an'}\,.
\end{aligned}
\tag{A4}
$$

Substituting these equations in Eq. (A3) and following the same procedure as in Scully and Lamb (Ref. 1) of making the Wigner-Weisskopf approximation and tracing over the reservoir variables, we obtain the Eqs. (3.2b) with

$$
\begin{aligned}
&\gamma_a = 2\pi W(\omega(ac))\,|\,V^a_{a0,c1}\,|^2\,,\\
&\gamma_b = 2\pi W(\omega(bd))\,|\,V^b_{b,0,d1}\,|^2\,,\\
&\gamma_c = 2\pi W(\omega(cd))\,|\,V^c_{c0,d1}\,|^2\,,\\
&\gamma_f = 2\pi W(\omega(fl))\,|\,V^f_{f0,l1}\,|^2\,,\\
&\gamma_m = 2\pi W(\omega(am))\,|\,V^m_{a0,m1}\,|^2\,,\\
&\gamma_{ab} = \tfrac{1}{2}(\gamma_a+\gamma_b+\gamma_m)\,,\\
&\gamma_{cf} = \tfrac{1}{2}(\gamma_c+\gamma_f+\gamma_a)\,.
\end{aligned}
$$

Here the W's are the corresponding phase-space mode densities for the $a\to c$, $b\to d$, $c\to d$, $f\to l$, and $a\to m$ transitions.[1]

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