Laser with a Fluctuating Pump: Intensity Correlations of a Dye Laser

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A new, first-principles derivation of the equations for a quantized single-mode laser field pumped by a semiclassical stochastic field in a four-level molecular pumping scheme is presented. Computer simulations are used to generate fits to measured correlation functions for a single-mode dye laser.

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Striking differences in the noise properties of dye lasers as compared to helium-neon lasers were recently revealed in measurements of the photon statistics and intensity correlation functions of a single-mode dye laser, performed by Kaminishi *et al.*[1] These measurements, and subsequent theoretical analyses,[2–4] have shown the importance of incorporating fluctuations in the pump source. The inclusion of pump fluctuations has been shown to introduce vastly different behavior of the laser output from what would be expected from the conventional quantum theory of the laser.[5–9] The inadequacy of the theoretical approaches[2–4] taken till now arises from the *ad hoc* inclusion of noise terms in the semiclassical equations of motion for the laser intensity. A consistent, comprehensive theory is clearly required to understand the correct form of the noise sources, and to derive their effects on the intensity fluctuations of the laser. It is the purpose of this note to report a new, first-principles theory for a laser pumped by a stochastic source.

Kaminishi *et al.*[1] attempted to apply Risken's theory[7,8] for single-mode, conventional lasers near threshold to their measurements on the dye laser. The theory is a Langevin approach to laser noise, governed by the Lamb semiclassical equation enhanced with an additive noise source:

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

in which E is the complex amplitude of the laser field, a and A are the pump parameter and saturation coefficient for the laser medium, and $\xi(t)$ is a Gaussian, white-noise source. Since the Risken theory does not explain the measured variations of the relative intensity fluctuations $[\langle(\Delta I)^2\rangle/\langle I\rangle^2]$ with pumping, or the observed correlation functions $[\lambda(\tau) = \langle \Delta I(t)\Delta I(t+\tau)\rangle/\langle I\rangle^2]$, Kaminishi *et al.* proposed that the pump parameter was not a constant but a fluctuating quantity. Their efforts were confined to fitting the $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ vs a curve.

Graham, Höhnerbach, and Schenzle[2] pursued this idea further by assuming

$$a = a_0 + \xi(t), \tag{2}$$

where a_0 is an average pump parameter. $\xi(t)$ was taken to be Gaussian, white noise. Neglecting the additive spontaneous-emission noise term, and the fact that A should also contain pump fluctuations, they obtained analytic solutions[10,11] of the Langevin equation with multiplicative noise.[12] They fitted the data concerning the relative intensity fluctuations as well as one correlation function of the dye-laser intensity. Unfortunately, this analysis did not fit some unpublished data, for other operating points of the laser, as was pointed out by Short, Mandel, and Roy in a more recent paper,[3] who also suggested the use of colored noise.

With colored noise, the equation

$$\dot{E} = [a_0 - A|E|^2]E + E\xi(t) \tag{3}$$

is no longer analytically solvable. A simulation of (3) with a stochastic pump parameter a was performed by Dixit and Sahni.[4] They achieved some success in explaining the observed data of Kaminishi *et al.*[1] and Short, Mandel, and Roy[3] by using colored noise for the pump. The results of our new, first-principles theory for lasers with fluctuating pumps unambiguously determines the location of noise terms as well as their detailed structure. Moreover, the theory contains spontaneous-emission contributions.

We use a density-matrix description of the active molecular energy levels and the quantum state of the laser field. The pump field is treated semiclassically. The active molecule is treated as a four-level system, with decay rates γ_{ij}.

The density-matrix equation of motion is

$$i\hbar\, \partial\rho/\partial t = [H,\rho], \tag{4}$$

where ρ is the density matrix for both the molecular levels and the photon states and H is the total

Hamiltonian,

$$H=\hbar\omega_{23}a^\dagger a+\sum_{i=1}^{4}|i\rangle\epsilon_i\langle i|+\mu\cos(\omega_{14}t)[|4\rangle E_p(t)\langle 1|+|1\rangle E_p^*(t)\langle 4|]$$
$$+g(a+a^\dagger)(|3\rangle\langle 2|+|2\rangle\langle 3|)+H_{\rm decay}, \quad (5)$$

with $H_{\rm decay}$ denoting Hamiltonians for decays.

The first term is the Hamiltonian for the quantized single-mode laser field with frequency ω_{23}. The second term is the Hamiltonian for the molecular energy levels with energy ϵ_i. The third term is for the pump field $E_p(t)$ interacting resonantly with levels 1 and 4. The electric dipole coupling strength is μ. $E_p(t)$ is a possibly stochastic, semiclassical field, allowing for noisy fluctuations in the pump laser source. The fourth term is for the interaction between the lasing energy levels (2 and 3) and the quantized laser field (single mode). The coupling coefficient is g. The remaining terms are stochastic Hamiltonians representing nonradiative transitions and cavity decay.

The method devised by Fox[12] has been used to obtain the equations of motion for the total system density-matrix elements. A contraction of this description is now possible which utilizes adiabatic elimination of off-diagonal molecular level matrix elements. We obtain, with $\rho=\exp\{(i/\hbar t)[H_0,\cdot]\}\hat\rho$ and $\bar\mu=\mu/\hbar$, $\bar g=g/\hbar$,

$$\partial\hat\rho_{11}/\partial t=-\bar\mu^2\int_0^t ds\exp[-\gamma_{14}(t-s)/2][E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]\hat\rho_{11}(s)$$
$$+(\gamma_{12}\hat\rho_{22}+\gamma_{13}\hat\rho_{33})+\mathscr{D}\hat\rho_{11}, \quad (6)$$

$$\partial\hat\rho_{22}/\partial t=-\gamma_{12}\hat\rho_{22}+\gamma_{23}\hat\rho_{33}$$
$$+\bar g^2\int_0^t ds\exp[-\gamma_{23}(t-s)/2][a^\dagger\hat\rho_{33}(s)a-\hat\rho_{22}a^\dagger a-a^\dagger a\hat\rho_{22}(s)-a^\dagger\hat\rho_{33}(s)a]+\mathscr{D}\hat\rho_{22}, \quad (7)$$

$$\partial\hat\rho_{33}/\partial t=-(\gamma_{13}+\gamma_{23})\hat\rho_{33}+\bar\mu^2\int_0^t ds\exp[-\gamma_{14}(t-s)/2]\ [E_p(t)E_p^*(s)+E_p^*(t)E_p(s)]\hat\rho_{11}(s)$$
$$+\bar g^2\int_0^t ds\exp[-\gamma_{23}(t-s)/2][a\hat\rho_{22}a^\dagger-\hat\rho_{33}(s)aa^\dagger-aa^\dagger\hat\rho_{33}(s)+a\hat\rho_{22}(s)a^\dagger]+\mathscr{D}\hat\rho_{33}. \quad (8)$$

The equation for the field density matrix ($\hat\sigma\equiv\hat\rho_{11}+\hat\rho_{22}+\hat\rho_{33}$) is

$$\partial\hat\sigma/\partial t=\mathscr{D}\hat\sigma+\bar g^2\int_0^t ds\exp[-\gamma_{23}(t-s)/2][2a^\dagger\hat\rho_{33}(s)a-\hat\rho_{33}(s)aa^\dagger-aa^\dagger\hat\rho_{33}(s)$$
$$+2a\hat\rho_{22}(s)a^\dagger-\hat\rho_{22}(s)a^\dagger a-a^\dagger\hat\rho_{22}(s)]. \quad (9)$$

Here $\mathscr{D}$ represents the cavity decay with

$$\mathscr{D}=\textstyle\sum_n(-n\lambda|n\rangle\langle n|\cdot|n\rangle\langle n|+(n+1)\lambda|n\rangle\langle n+1|\cdot|n+1\rangle\langle n|), \quad (10)$$

where $|n\rangle$ is the n-photon state of the laser field and λ is the cavity decay rate. Taking photon state matrix elements of these equations and performing the trace of them yields, after adiabatic elimination of the population in level 2, a much reduced description given by the pair of equations

$$\dot N_3=-(\gamma_{13}+\gamma_{23})N_3+\bar\mu^2\int_0^t ds\exp[-\gamma_{14}(t-s)/2][E_p(t)E^*(s)+E_p^*(t)E_p(s)][N-N_3(s)]$$
$$-2\bar g^2\int_0^t ds\exp[-\gamma_{23}(t-s)/2]N_3(s)[I(s)+1], \quad (11)$$

$$\dot I=-\lambda I+2\bar g^2\int_0^t ds\exp[-\gamma_{23}(t-s)/2]N_3(s)[I(s)+1], \quad (12)$$

where we have defined $\sum_n N\langle n|\hat\rho_{33}|n\rangle\equiv N_3$ and $\sum_n\langle n|a^\dagger a\hat\rho|n\rangle=I$ in which I is the laser intensity and N_3 is the population of the excited lasing level of the molecule.

If the pump field fluctuations are very fast, one may make the instantaneous approximation,[13] and replace the memory integrals by instantaneous terms, to obtain

$$\dot{N}_3 = G(t) - \lambda'(t) N_3 - BN_3(I+1), \tag{13}$$

$$\dot{I} = -\lambda I + BN_3(I+1). \tag{14}$$

in which

$$G(t) = (4\bar{\mu}^2/\gamma_{14}) N I_p(t),$$

$$\lambda'(t) = (\gamma_{13} + \gamma_{23}) + (4\bar{\mu}^2/\gamma_{14}) I_p(t),$$

$$B = 2\bar{g}^2/\gamma_{23}.$$

$I_p(t) = [I_{p0}^{1/2} + \xi(t)]^2$ is the quantity which may fluctuate. I_{p0} is the average pump intensity; $\xi(t)$ is Gaussian, white noise. Adiabatic elimination of N_3 by

$$N_3 = \frac{G}{\lambda' + B(I+1)} \tag{15}$$

leads to

$$\dot{I} = -\lambda I + \frac{G(t)\, B(I+1)}{\lambda'(t) + B(I+1)} \tag{16}$$

which represents the strong-signal version of Eq. (3). We should note several important differences at this point. Both $G(t)$ and $\lambda'(t)$ contain $I_p(t)$, which is stochastic. The 1 in the $I+1$ terms is a result of including spontaneous emission in a systematic manner in our equations. If one contemplates using colored noise for $I_p(t)$, one should actually use Eqs. (11) and (12) with the correct form of the memory integrals. Moreover, the decay rate γ_{14} must be compared with the decay rate of the autocorrelation function for E_p to determine which dominates the value of the integral over the past in Eq. (11).

From a stochastic point of view, Eqs. (13) and (14) are more easily studied than Eq. (16), because the pump fluctuations enter both G and λ' in Eq. (16).

We have carried out computer simulations of Eqs. (11) and (12) with the assumption that the fluctuations of the multimode pump laser (which may have well over 50 axial modes) are very fast compared to other time scales of the molecular processes. Correlation functions $\lambda(\tau) = \langle \Delta I(t) \times \Delta I(t+\tau)\rangle / \langle I \rangle^2$ have been obtained from the simulations, in which the pump field is assumed to contain Gaussian, white noise. A large number of parameters occur in Eqs. (11) and (12). We have made plausible estimates for the parameters of the dye molecule (Rhodamine 6G) and obtained fits for the three intensity correlation functions of which two are shown here [Figs. 1(a) and 1(b)]. These were experimentally measured for different operating points of a single-mode dye laser.[3] The fits have been obtained by varying the average pump intensity and the coefficient B, i.e., *three curves have been fitted by varying two parameters.* The time step size taken was 1 ns and 300 realizations of 40 000 steps each were computed. Other parameters are

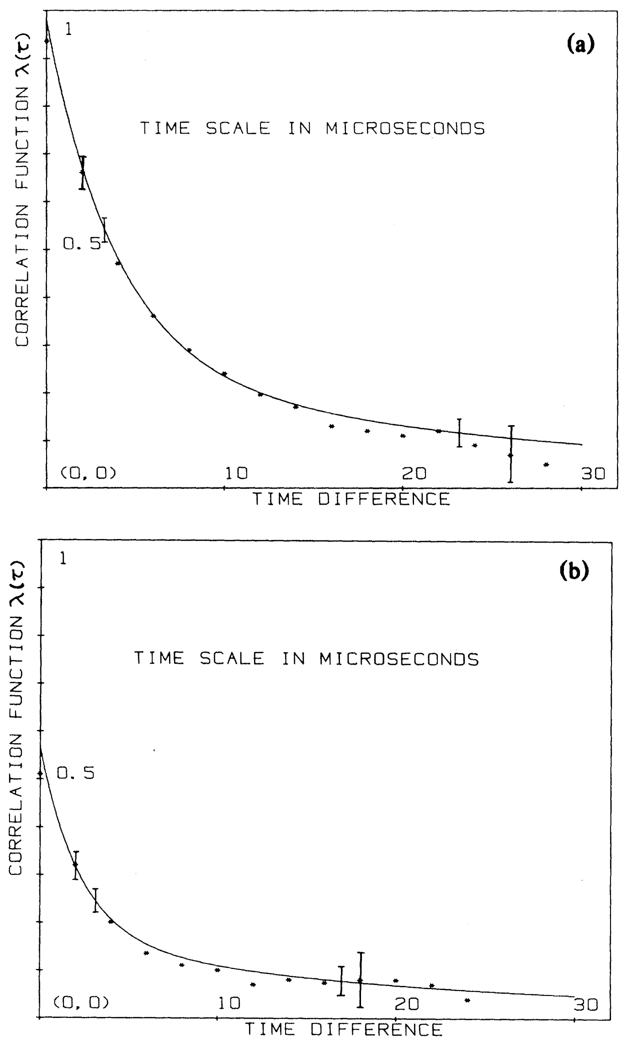

FIG. 1. $\lambda(\tau)$ vs τ. Solid line, experimental data from Ref. 3; asterisks, simulation results. The error bars are estimated from the scatter of the experimental points and the results of the simulation for different realizations. (a) $\lambda(0)_{\rm exp} = 0.98$; $I_{p0} = 2.0 \times 10^9$; $B = 0.0033\ \rm sec^{-1}$. (b) $\lambda(0)_{\rm exp} = 0.57$; $I_{p0} = 2.4 \times 10^9$; $B = 0.0037\ \rm sec^{-1}$. The other parameters are common to both figures: $8A/\gamma_{14} = 10^{-4}\ \rm sec^{-1}$, $\gamma_{14} = 1.4 \times 10^5\ \rm sec^{-1}$, $\lambda = 5 \times 10^5\ \rm sec^{-1}$, $Q = 7.0 \times 10^2$, $N = 10^{10}$ molecules, $\gamma_{13} + \gamma_{23} = 10^7\ \rm sec^{-1}$, single integration time step $= 10^{-9}$ sec.

stated in the figure captions, and the estimated errors in the experimental and simulation results are shown in the figures. No "subtraction" procedures have been used.[2-4]

It is also easy to see the effect of the pump fluctuations on the quantum mechanical equations of motion for the reduced field density matrix. On taking $\langle n|\cdot|n\rangle$ matrix elements of Eqs. (6)–(9), neglecting the population of level 2, and adiabatically eliminating $\rho_{33}(n)$ ($\equiv \langle n|\rho_{33}|n\rangle$), we obtain

$$\dot{\hat{\sigma}}(n) = -n\lambda\hat{\sigma}(n) + (n+1)\lambda\hat{\sigma}(n+1) + \frac{\bar{g}^2}{\frac{1}{2}\gamma_{23}(\gamma_{13}+\gamma_{23})}\left[\frac{nG(t)\hat{\sigma}(n-1)}{1+\bar{g}n^2/\frac{1}{2}\gamma_{23}(\gamma_{13}+\gamma_{23})} - \frac{(n+1)G(t)\hat{\sigma}(n)}{1+\bar{g}^2(n+1)/\frac{1}{2}\gamma_{23}(\gamma_{13}+\gamma_{23})}\right]. \quad (17)$$

Here

$$G(t) = \bar{\mu}^2\int_0^t ds\,\exp[\gamma_{14}(t-s)/2][E_p(t)E_p^*(s) + E_p^*(t)E_p(s)]. \quad (18)$$

This equation has nearly the same form as the Scully-Lamb master equation.[14]

The theory presented here provides a comprehensive, consistent approach which considers from first principles the inclusion of pump noise in a laser. The details of this theory as well as extensions will be published in a forthcoming paper.[13]

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