[3]G. Herzberg, Nature (London) 166, 563 (1950).
[4]M. Trefler and H. P. Gush, Phys. Rev. Lett. 20, 703 (1968).
[5]W. Kolos and L. Wolniewicz, J. Chem. Phys. 45, 944 (1966).
[6]P. R. Bunker, J. Mol. Spectrosc. 46, 119 (1973).
[7]A. L. Ford and J. C. Browne, Phys. Rev. A 7, 418 (1973).
[8]L. Wolniewicz, Can. J. Phys. 53, 1207 (1975).
[9]L. Wolniewicz, Can. J. Phys. 54, 672 (1976).
[10]A. L. Ford and J. C. Browne, Phys. Rev. A 16, 1992 (1977).
[11]H. L. Welsh, in *Spectroscopy*, edited by D. A. Ramsey, MTP International Review of Science, Physical Chemistry Series, Vol. 3 (Univ. Park Press, Baltimore, 1972), p. 33.
[12]J. D. Poll, R. H. Tipping, R. D. G. Prasad, and S. P. Reddy, Phys. Rev. Lett. 36, 248 (1976).
[13]R. H. Tipping, J. D. Poll, and A. R. W. McKellar, Can. J. Phys. 56, 75 (1978).
[14]R. M. Herman, R. H. Tipping, and J. D. Poll, Phys. Rev. A 20, 2006 (1979).
[15]The absorption coefficient $\alpha(\omega) = (1/\rho l) \ln[I_0(\omega)/I(\omega)]$. $I_0(\omega)$ is the density transmitted in the absence of the absorber and $I(\omega)$ is the intensity transmitted through the absorber. l is the path length and ρ is the gas density. Amagat units are used here for density.
[16]U. Fano, Phys. Rev. 124, 1866 (1961).

Statistical Properties of Light from a Dye Laser

R. Graham, M. Höhnerbach, and A. Schenzle
Fachbereich Physik, Universität Essen, D-4300 Essen, West Germany
(Received 25 March 1982)

Recent experimental results of Kaminishi *et al.* on the photon statistics of a dye laser are compared with the exact solution of a laser model with fluctuating pump parameter.

PACS numbers: 42.55.Mv

One of the great successes of quantum optics was the theoretical derivation of the statistical properties of laser light and the subsequent detailed experimental confirmation. The laser was thereby established as a source of light with statistical properties fundamentally different from all thermal light sources.

The well-known theoretical model which was successful in explaining all details of the photon statistics of a single-mode laser near threshold is the simple Van der Pol oscillator in rotating wave approximation[1]:

$$\dot{\beta} = [(a_1 + ia_2) - (A_1 + iA_2)|\beta|^2]\beta + \xi(t). \qquad (1)$$

Here $\beta(t)$ is the complex amplitude of the laser mode, a_1, a_2, A_1, and A_2 are real parameters, a_2 and A_2 are different from zero only for nonzero detuning, a_1 is the pump parameter and positive (negative) above (below) threshold, and $A_1 > 0$ provides for stabilization above threshold due to saturation. $\xi(t)$ in Eq. (1) is a Gaussian white-noise source with the properties $\langle \xi(t) \rangle = 0$, $\langle \xi(t) \times \xi(0) \rangle = 0$, $\langle \xi^*(t)\xi(0) \rangle = Q\delta(t)$, which turns Eq. (1) into a stochastic differential equation. Henceforth, we always use the Stratonovich calculus of such equations.

Equation (1) should apply to any single-mode laser, and is in this sense universal, provided that the laser is sufficiently close to its threshold, where corrections due to higher-order nonlinearities or to time derivatives and deviations of $\xi(t)$ from white noise become irrelevant. In particular, the form of Eq. (1) is independent of any microscopic details of the laser process, and already follows from general symmetry principles of the theory of continuous instabilities.[2]

Recently, Kaminishi *et al.*[3] reported very interesting experimental results on the photon statistics of a dye laser near threshold. Their results differ completely from what one would expect on the basis of model (1). For instance, in Ref. 3 it was found that the relative mean square of the intensity fluctuations $\langle \Delta I^2 \rangle / \langle I \rangle^2$ increases up to values of ~1000 for sufficiently small average photon number $\langle n \rangle$, whereas model (1) predicts an upper bound $\langle \Delta I^2 \rangle / \langle I \rangle^2 \leq 1$. The model which adequately describes a dye laser near threshold must therefore differ from Eq. (1) by terms which do not become irrelevant, even very close to the laser threshold.

In the present Letter we propose such a model, which turns out to be exactly solvable, and compare its predictions with the available experimental data. The good agreement which we find supports the idea that the dye-laser threshold belongs to the new class of continuous instabilities with fluctuating control parameter which is described, again in a universal way, by our model.

More experimental work along the lines of Ref. 3, which we hope to stimulate by the results presented here, would allow tests of the very detailed quantitative predictions of the model.

The model we wish to propose in order to describe a dye laser near threshold differs from Eq. (1) in that the pumping parameter a_1 and the frequency shift a_2 are both subject to rapid time-dependent fluctuations around some fixed mean values $a_{1,0}$, $a_{2,0}$. Such fluctuations in gain and refractive index naturally occur in a dye laser as a result of turbulence and concentration fluctuations in the dye cell. In fact, Kaminishi *et al.*[3] already interpreted some of their results by assuming fluctuations of the gain to be present, an idea which we adopt and strengthen in this Letter. However, these authors did not construct a dynamical model incorporating such fluctuations and therefore did not attempt to interpret the dynamics of the observed intensity fluctuations of the laser light in terms of this idea.

Close to the laser threshold the fluctuations of the light field are known to slow down. In our model we assume that on this long time scale the rapid fluctuations of gain and refractive index may be considered as Gaussian white noise. Assuming for simplicity that these fluctuations are more important than the noise due to spontaneous emission, we arrive at the equations of model (2), given by (1) with $\xi(t)\equiv 0$, and

$$\langle[a_i(t)-a_{i,0}][a_j(0)-a_{j,0}]\rangle = Q_{ij}\delta(t). \tag{2}$$

The model (2) has been solved exactly in earlier papers.[4-6] The time dependence of the $a_i(t)$ does not become irrelevant as the threshold $a_{1,0}=0$ is approached. For example, the critical exponent of $\langle|\beta|\rangle$ at threshold in the steady state is changed from $\frac{1}{2}$ for $Q_{ij}=0$ to 1 for $Q_{ij}\neq 0$.[6] Here we want to compare the predictions of the model with the experimental results of Ref. 3 on the photocount distribution, the relative mean square of the intensity fluctuations as a function of the average photon number, and the intensity correlation function in the steady state.

The time-independent distribution $W_0(I)$ of the intensity $I=|\beta|^2$ is given by

$$W_0(I)=\begin{cases}\delta(I), & \alpha\leq 0,\\ [q^\alpha/\Gamma(\alpha)]I^{\alpha-1}\exp(-qI), & \alpha>0,\end{cases} \tag{3}$$

with $q=Q_{11}/4A_1$, $\alpha=a_{1,0}/Q_{11}$. Small additive fluctuations described by $\xi(t)$ in Eq. (1) would smear out the δ function obtained for $\alpha\leq 0$ but would not affect the result for $\alpha>0$ in an appreciable way.[4] In the following, only the results for $\alpha>0$ will be needed.

The photocount distribution associated with (3) is given by[7]

$$p(n)=(1/n!)\int_0^\infty dI\, W_0(I)(\gamma I)^n e^{-\gamma I}, \tag{4}$$

where γ is the counting time ($\sim 1\ \mu$sec) times a constant, characteristic of the detector. We obtain explicitly

$$p(n)=\begin{cases}\delta_{n,0}, & \alpha\leq 0,\\ \dfrac{(1-C)^\alpha}{\Gamma(\alpha)}\dfrac{\Gamma(\alpha+n)}{n!}C^n, & \alpha>0,\end{cases} \tag{5}$$

with $C=\gamma/(\gamma+q)$. The pump parameter α is related to the average photon number by

$$\langle n\rangle=\begin{cases}0, & \alpha\leq 0,\\ \alpha C/(1-C), & \alpha>0,\end{cases} \tag{6}$$

and to the relative mean square of the intensity

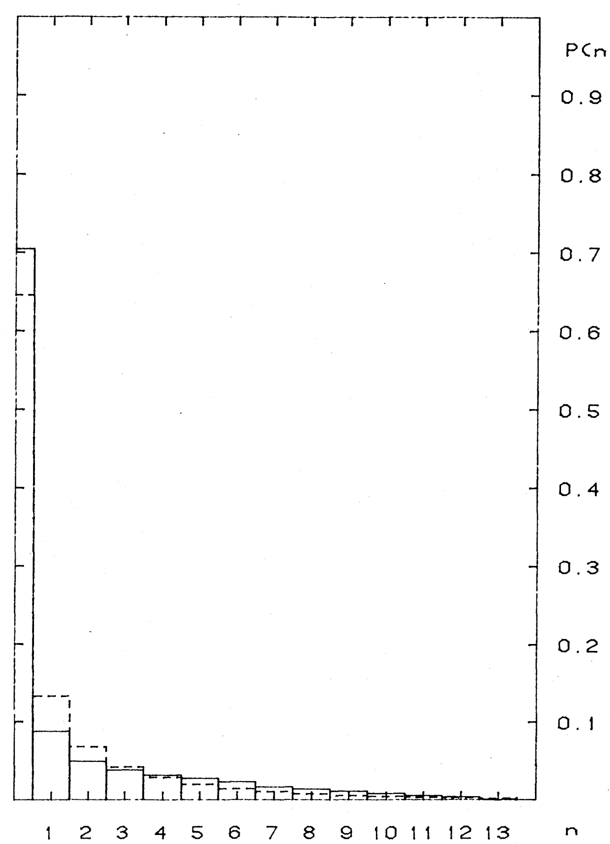

FIG. 1. Photocount distribution. Full line, experimental result from Ref. 3; broken line, theoretical result Eq. (5) by fitting C.

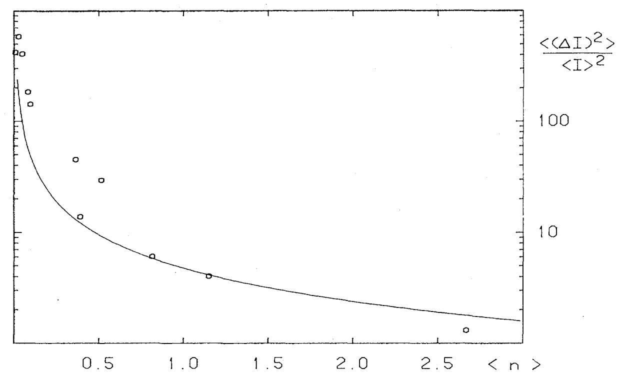

FIG. 2. Relative mean square of intensity fluctuations, Eqs. (6) and (7), with C determined from Fig. 1. Experimental points are from Ref. 3.

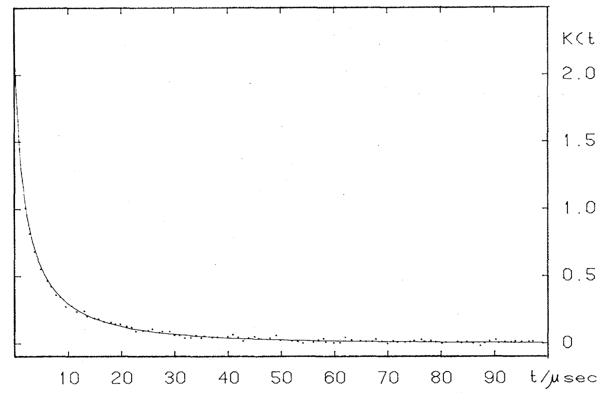

FIG. 3. $K(t)$ from Eq. (8) for $\alpha=\frac{1}{2}$ by fitting Q_{11}. Experimental points are obtained from Ref. 3 after subtraction of a constant background.

fluctuations by

$$\langle\Delta I^2\rangle/\langle I\rangle^2=\alpha^{-1}. \tag{7}$$

The photocount distribution (4) was measured in Ref. 3 for $\langle\Delta I^2\rangle/\langle I\rangle^2=4$ which implies $\alpha=\frac{1}{4}$ by (7). In Fig. 1 we present a comparison with (5), where C is determined by fitting.

The model is seen to describe remarkably well the strong peaking of the photocount distribution at zero photon number, which is qualitatively different from the photocount distribution of a usual laser near threshold, and caused by the fluctuations of the gain.

The relative mean square of the intensity fluctuations was also measured in Ref. 3 as a function of $\langle n\rangle$. Since C is already determined by the previous fit, Eqs. (6) and (7) may now be confronted with experiment without any further fitting. The result is shown in Fig. 2. In view of the considerable scatter of the experimental points, there is reasonably good agreement. This lends further support to the result (5) from which C was derived, and to the model (2).

The intensity correlation function[8,9] $K(t)=\langle\Delta I(t)\times\Delta I(0)\rangle/\langle I\rangle^2$ of the model (2) has been determined in Refs. 5 and 6 up to quadrature by exactly solving the spectral problem of the associated Fokker-Planck equation. In the domain $\alpha<2$ of parameter space, the Fokker-Planck spectrum was found to consist of a pure continuum, which led to the representation

$$K(\tau)=\exp(-\alpha^2\tau)\int_{-\infty}^{\infty}d\kappa\,\frac{(\alpha^2+\kappa^2)\kappa\sinh(\pi\kappa)\Gamma(\alpha+i\kappa/2)\exp(-\kappa^2\tau)}{4\Gamma(\alpha)[\cosh(\pi\kappa)-\cos(\pi\alpha)]} \tag{8}$$

with $\tau=\frac{1}{2}Q_{11}t$. The intensity correlation function was measured in Ref. 3 for $\langle\Delta I^2\rangle/\langle I\rangle^2=2$, i.e., $\alpha=\frac{1}{2}$ by Eq. (7). The attempt to compare directly with Eq. (8) meets with the following difficulty: The experimental result yields a $K(t)$ that decays in about 30 μsec to a finite background value, which then decays only on a much longer time scale,[3] and may therefore be considered as constant on the time scale of the initial decay.

Since the correlation function $K(t)$, in any case, must be extracted from the measurements by the judicious subtraction of a time-independent background,[10] we have chosen, differently from Ref. 3, to include this constant background in the subtraction before comparing with Eq. (8). The comparison, shown in Fig. 3, is then made by fitting Q_{11}, the only parameter left. The experimental curve is found to be in excellent agreement with the nonexponential decay as given by Eq. (8). The parameters Q_{11} and γA_1 of the model are now fixed and given by $Q_{11}=0.18$ $(\mu\text{sec})^{-1}$ and $\gamma A_1/Q_{11}=1.18$.

In conclusion we note that our model is able to explain from a unified point of view the available data on the statistics and the dynamics of light from a dye laser near threshold. The model has only two adjustable parameters, as far as phaseless quantities like photon number and intensity are concerned, just as in the highly successful model (1). Experimental results for the same system with different values of the pump parameter are now predicted by the results of Refs. 5 and 6 without further adjustable parameters, and can be used to test the model further.

[1]H. Risken, in *Progress in Optics*, edited by E. Wolf (North-Holland, Amsterdam, 1970), Vol. VIII, p. 239.

[2]R. Graham, *Springer Tracts in Modern Physics* (Springer, Berlin, 1973), Vol. 66, p. 1.

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

[4]A. Schenzle and H. Brand, Phys. Rev. A 20, 1628 (1979).

[5]R. Graham and A. Schenzle, Phys. Rev. A 25, 1731 (1982).

[6]R. Graham, to be published.

[7]L. Mandel, Proc. Phys. Soc. London 72, 1037 (1958), and 74, 233 (1959).

[8]M. Born and E. Wolf, *Principles of Optics* (Pergamon, Oxford, 1975).

[9]L. Mandel and E. Wolf, Phys. Rev. 124, 1696 (1961).

[10]S. Chopra and L. Mandel, Rev. Sci. Instrum. 43, 1489 (1972).