Photon Statistics of a Dye Laser Far Below Threshold

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The relative mean square fluctuations $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ of the intensity I of a single-mode dye laser have been measured by a photon counting technique, in the region from about threshold to intensities 1000 times below threshold. The results show a steady increase of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ from less than 1 to about 150 as $\langle I\rangle$ is reduced, followed by a rapid drop to zero. This behavior appears to be described fairly well by an equation of motion containing both additive spontaneous emission fluctuations and multiplicative pumping fluctuations.

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The dye laser has proved to be not only a valuable tool for experimental work because of its tunability, but also a laser that is unusually interesting in its own right. Recent experimental work[1-3] and its subsequent theoretical analysis[4-9] suggest that the pump parameter a in the usual Lamb equation of motion[10] for the laser field has to be treated as a random function $a(t)$. As $a(t)$ multiplies the field amplitude $E(t)$ in the equation of motion, this implies that we have a multiplicative type of noise. The quantum or spontaneous-emission noise, on the other hand, which is usually treated as additive, has sometimes been left out of the equation, because its magnitude is so much smaller.

We have recently obtained strong indications that the additive quantum noise nevertheless plays an essential role in a two-mode dye laser well above threshold,[3] because it is responsible for the observed mode switching, which is suppressed in its absence. We now wish to present some new experimental results on a single-mode laser operating below threshold, which also demonstrate the importance of spontaneous emission noise.

Figure 1 shows an outline of the apparatus. The active laser medium is rhodamine 6G, which is made to flow at high speed through a cell, and is optically pumped by the light of an argon ion laser. Three etalons ensure single-mode operation at a wavelength of about 6000 Å. The light emerging from the output mirror is attenuated and then falls on a beam splitter, where it is split into two beams that are directed to two counting photomultiplier tubes. After amplification and pulse shaping, the photoelectric pulses from the two detectors are counted, and also fed to the two inputs of a coincidence counter, that registers the coincident arrival of pulses within its resolving time T_R. When corrections for background are included, the rate of counting $\mathfrak{R}$ of the coincidence counter is given by

$$\mathfrak{R} = R_1 R_2 T_R\left[1 + \frac{1}{T_R}\int_{-T_R/2}^{T_R/2} d\tau\, \lambda(\tau) + \frac{r_1}{R_1} + \frac{r_2}{R_2} + \frac{r_1 r_2}{R_1 R_2}\right]. \tag{1}$$

Here R_1, R_2 are the average counting rates of the two detectors attributable to the laser, r_1, r_2 are background rates, and

$$\lambda(\tau) \equiv \langle \Delta I(t)\, \Delta I(t+\tau)\rangle/\langle I\rangle^2 \tag{2}$$

is the normalized autocorrelation function of the fluctuations of the light intensity $I(t)$. When T_R is much shorter than the natural intensity correlation time T_c, then $\lambda(\tau)$ can be replaced by $\lambda(0)$ to a good approximation under the integral, and Eq. (1) reduces to

$$\mathfrak{R} = R_1 R_2 T_R\left[1 + \lambda(0) + \frac{r_1}{R_1} + \frac{r_2}{R_2} + \frac{r_1 r_2}{R_1 R_2}\right]. \tag{3}$$

$\lambda(0)$ is also the normalized second factorial moment $\langle n(n-1)\rangle/\langle n\rangle^2 - 1$ of the number of photon counts n in a short time interval. It is clear from this equation that measurements of $\mathfrak{R}$ allow $\lambda(0)$ to be determined. In practice the value of T_R is governed by the discriminator pulse length, which was of order 18 nsec. At least in the thresh-

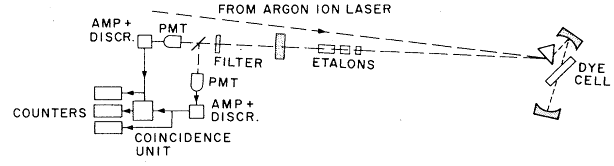

FIG. 1. Outline of the apparatus.

old region and above, T_R is generally much less than typical intensity correlation times, that usually range from microseconds to milliseconds.

Figure 2 shows the experimental results of a determination of the relative mean square intensity fluctuation $\lambda(0) = \langle(\Delta I)^2\rangle/\langle I\rangle^2$ from Eq. (3), as a function of the mean light intensity $\langle I\rangle$. The standard deviations of $\lambda(0)$ either are indicated or are smaller than the dot size. As the working point of the laser is lowered below threshold, $\lambda(0)$ rises from small values to a maximum of about 150, and it then drops rather rapidly close to zero. The value $\lambda(0) = 0.57$ represents the threshold for a conventional laser, so that we may characterize our measurements as being concentrated in the region well below threshold. However, as has long been known, the dye laser is very poorly described by conventional laser theory,[1-3] according to which $\lambda(0)$ should never rise above unity.

It was suggested a few years ago[2] that the departures of the behavior from conventional laser theory may be attributable to pumping fluctuations, and the treatment of the laser pump parameter as a random variable does indeed lead to the prediction that $\lambda(0)$ should rise with decreasing $\langle I\rangle$ below threshold. More recently this idea has been developed into a dynamical theory by Graham, Schenzle, and their co-workers.[4-6,9] They took the dimensionless equation of motion for the complex laser field $E(t)$ to be of the general form

$$\dot{E}(t) = [a_0 + a_1(t) - |E(t)|^2]E(t) + q(t), \tag{4}$$

except that the additive noise term $q(t)$ representing the quantum fluctuations was left out. Here a_0 is the average pump parameter and $a_1(t)$ is a Gaussian random function representing the pumping fluctuations about the mean. Originally $a_1(t)$ was taken to be δ-correlated. The model was improved by Dixit and Sahni,[8] who took $a_1(t)$ to be an Ornstein-Uhlenbeck process with

$$\langle a_1^*(t)a(t+\tau)\rangle = Q\Gamma e^{-\Gamma|\tau|}, \tag{5}$$

and solved the equation of motion numerically. Schenzle and Graham later adopted the same model without spontaneous emission fluctuations as representative of a dye laser above threshold, and they succeeded in obtaining analytic solutions of the associated Fokker-Planck equation.[9] This led to the prediction that $\lambda(0)$ should be proportional to the inverse light intensity $1/\langle I\rangle$, which is close to what is observed except at the lowest intensities (see Fig. 2), and it also led to reasonably good agreement with experiment for the form of $\lambda(\tau)$.[4,8,9] The model does not predict the observed peak in $\lambda(0)$, but this occurs far below threshold, well outside the domain of validity of the theory.

The large increase in $\lambda(0)$ as the working point of the laser is reduced below threshold is clearly attributable to pumping fluctuations. Indeed in the region near the peak, the laser is off almost all the time, and turns on spontaneously only for brief periods. However, in order to make $\lambda(0)$ come down again at sufficiently low intensities, it is necessary to include the spontaneous-emission fluctuations, represented by $q(t)$ in Eq. (4), as well.

We have used a Monte Carlo procedure to solve Eq. (4) iteratively, with $q(t)$ in the form of a complex δ-correlated Gaussian noise, scaled so that

$$\langle q^*(t)q(t+\tau)\rangle = 4\delta(\tau). \tag{6}$$

The results of the calculation with $Q = 300$ and $\Gamma = 5$ are also shown in Fig. 2, superimposed on the experimental data. Although these values of Q, Γ were chosen because they produced reasonable agreement with experiment, no systematic attempt was made to find the combination leading to the best fit. Also, the Monte Carlo procedure converges very slowly in the very low intensity region (at least for $\lambda(0) > 1$), and the accuracy of the theoretical curve is worst where it rises most rapidly from unity.

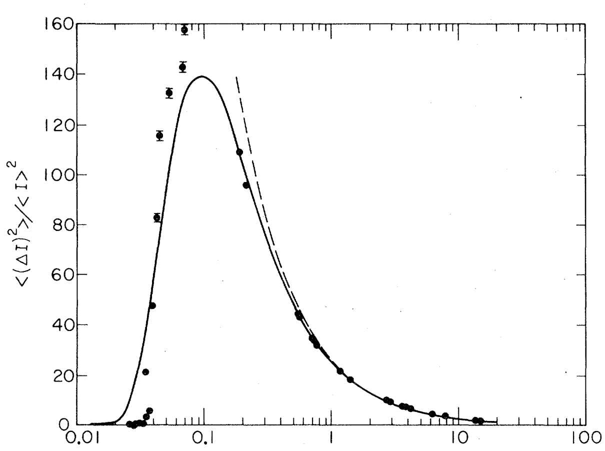

FIG. 2. Measured values of the relative fluctuations $\lambda(0) = \langle(\Delta I)^2\rangle/\langle I\rangle^2$, superimposed on the theoretical solution obtained from Eq. (4), with $Q=300$, $\Gamma=5$. The intensity scale is in arbitrary units. The broken curve is proportional to $1/\langle I\rangle$.

It will be seen that the general trend of the observed behavior appears to be quite well described by Eq. (4), except that experimentally $\lambda(0)\to 0$ as $\langle I\rangle \to 0$, whereas theoretically $\lambda(0)\to 1$, which represents the thermal limit. The difference may be connected with the fact that the linewidth can become so great far below threshold that Eq. (1) can no longer be approximated by Eq. (3), and the radiation field may behave as if it were effectively a multimode field. Although a peak in $\lambda(0)$ is also predicted by dye-laser theories that include the effects of crossover transitions between triplet states,[11,12] our results suggest that Eq. (4) is an adequate representation of the dye-laser field near threshold and below.

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