Photon statistics of a dye laser*

J. A. Abate, H. J. Kimble, and L. Mandel
Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627
(Received 8 March 1976)

The fluctuation properties of a cw dye laser, pumped by an argon-ion laser, have been investigated by photon-counting and two-time correlation measurements. The results show significant departures from the usual single-mode laser theory in the region of threshold and below. However, there are indications that the departures may be due to extraneous rather than intrinsic effects, and when these effects are subtracted out, the results are in substantial agreement with the predictions of the usual theory.

I. INTRODUCTION

The development of the tunable, continuous-wave dye laser[1] has provided us with a powerful tool that is increasingly being applied to the investigation of atomic and molecular systems. However, with few exceptions, the study of the dye laser itself, and particularly of its fluctuation properties, has received little attention.[2,3] In a recent theoretical treatment of the dye laser by Schaefer and Willis[4] it has been suggested that because of losses to the triplet states the statistical behavior of a dye laser may not follow the usual laser theory,[5,6] which has been confirmed in numerous measurements performed on He:Ne and other lasers,[7] under all working conditions. Moreover, because of the coherent optical pumping to which a cw dye laser is normally subjected, it may be expected to be especially sensitive to small pumping fluctuations when it is operating in the neighborhood of the oscillation threshold.

For all these reasons we felt it desirable to investigate the fluctuation and correlation properties of the light emitted by a dye laser by photoelectric counting techniques, for various working points of the laser. Our measurements indicate that there are indeed significant departures from conventional laser theory in the behavior of the dye laser, but that these departures appear to be connected with the manner in which the laser is normally operated rather than being fundamental. When suitable corrections for the extraneous effects are made, the measurements are in substantial agreement with the usual laser theory,[5,6] and no evidence for any significant departures is found.

II. EXPERIMENT

The dye laser used in these experiments was similar to one described by Schuda *et al.*[8] The optical arrangement is illustrated in Fig. 1. The dye, consisting of a soap and water solution of rhodamine 6G, was made to flow continuously through a pumping cell that was illuminated by 5145 Å light of a Spectra Physics 164 argon-ion laser. The ion laser was stabilized by its own internal feedback arrangement. At the same time, the long-term drift in the dye-laser output intensity was held to a fraction of 1% by another feedback loop, as previously described.[9] This second feedback loop allowed the working point of the dye laser to be controlled from well below to well above threshold. We found that by optically pumping with one spectral component (the 5145-Å line) of the argon laser, we were able to achieve improved output stability.

Single-mode operation of the dye laser was achieved through the insertion of three tilted glass etalons into the laser cavity. The dye-laser frequency could be varied over a wide range by tilting the output mirror shown in Fig. 1, and over a small range with the help of a piezoelectric crystal that moved the output mirror normally to its face. For the purpose of these experiments, the dye laser was operated at a wavelength of 5815 Å, and had a linewidth less than 1 MHz. In the course of one experimental run, the frequency suffered a typical thermal drift of about 150 MHz, which could be compensated by a frequency stabilization circuit when the laser was operated well above threshold.[9] However, in the threshold region, where most of our measurements were made, the frequency stabilization proved to be less satisfactory and was not used

The output beam from the dye laser was split into several beams with the aid of 45° half-silvered mirrors, as shown in Fig. 2, and two beams were allowed to fall on two photomultiplier tubes. The photoelectric pulses emitted by the detectors were amplified and shaped by discriminators, and were then sent via electronic gates to various counters under the control of a PDP 11/40 computer, as indicated in Fig. 2.

For the purpose of the fluctuation measurements, only one of the two detector channels was used. The number of pulses, n, received in a succession

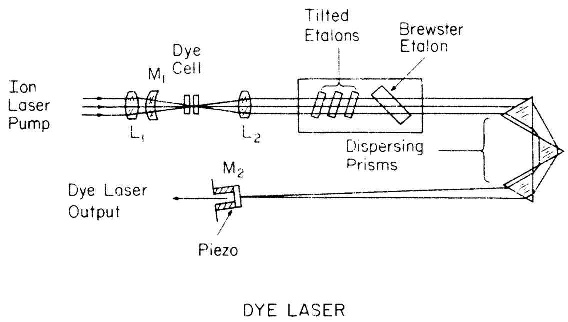

FIG. 1. Outline of the dye-laser system.

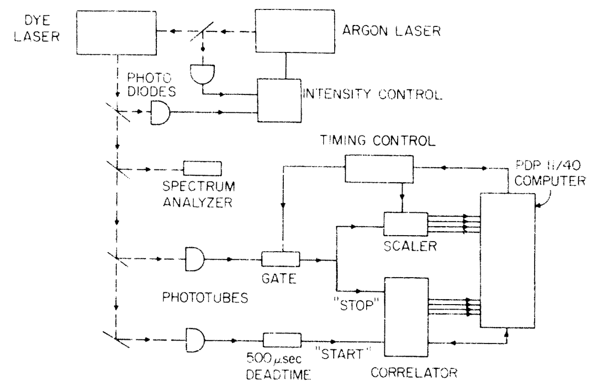

FIG. 2. Block diagram of the apparatus for photoelectric counting and two-time correlation measurements.

of 1-μsec-long counting intervals separated by 200–500 μsec was registered, and the information was stored in the memory of the computer in such a way that the number of times, N_n, that n events were counted could be directly displayed. This number provided a measure of the probability distribution $p(n)$, through the relation

$$p(n)=N_n\Big/\sum_{n=0}^{\infty} N_n\,, \tag{1}$$

for large N_n, from which the various moments or factorial moments of n can be calculated via the equations

$$\langle n^r\rangle=\sum_{n=0}^{\infty} n^r p(n)\,,\quad r=1,2,\cdots, \tag{2}$$

$$\langle n^{(r)}\rangle=\sum_{n=0}^{\infty} n(n-1)\cdots(n-r+1)p(n)\,,\quad r=1,2,\cdots\,. \tag{3}$$

The correlation measurements depend on the use of both photodetectors in two channels that we designate as the "start" pulse and the "stop" pulse channels, 1 and 2, respectively. As has been shown,[10] the joint twofold probability density of photoelectric detection, $P_2(t,t+\tau)$, by the two detectors at time t and at time $t+\tau$ is related to the instantaneous light intensities $I_1(t)$ and $I_2(t)$ at the two detectors by the formula

$$P_2(t,t+\tau)=q_1q_2c^2S_1S_2\langle I_1(t)I_2(t+\tau)\rangle\,, \tag{4}$$

where q_1 and q_2 are the quantum efficiencies and S_1 and S_2 the illuminated surface areas of the two detectors. The correlation function in Eq. (4) has been written as a classical correlation, but the same expression would hold in quantum electrodynamics if it were written in normal order. It follows that we can determine the intensity correlation function by recording the distribution of arrival times τ of stop pulses following the receipt of a start pulse. A digital technique for automatically doing so has previously been described.[11] We have improved it by placing the correlator under the control of the computer, and incorporating a number of computer checks to test the stability of the system and the validity of the data being received. Counting data that showed a drift of the mean light intensity of more than 1% were automatically discarded.

A number of corrections to the moments calculated from Eqs. (2) or (3) need to be made before the results can be compared with theoretical predictions. In the first place, a correction has to be made for the dead time of the counting electronics following each count. This results in corrections to the measured factorial moments $\langle n^{(r)}\rangle$ given by Eq. (3), which have been calculated by Chang *et al.*,[7] and were expressed by them as a power series in the ratio δ of the dead time to the counting-time interval. Secondly, allowance has to be made for photoelectric counts contributed by background light and by thermionic and other emissions from the photocathode. We measured the contribution from these sources directly by keeping the pumping light intensity constant and tilting one of the etalons until the dye laser was extinguished and only the fluorescence remained. If $p_B(n)$ is the probability distribution for counting n pulses due to these unwanted sources, and $p_L(n)$ the corresponding probability distribution for the light from the dye laser alone, then the measured probability distribution $p(n)$, containing contributions from both sources, is related to $p_L(n)$ and $p_B(n)$ to a first approximation by

$$p(n)=\sum_{m=0}^{n} p_L(m)p_B(n-m)\,. \tag{5}$$

The required probability $p_L(n)$ can therefore be extracted from the measured quantities $p(n)$, $p_B(n)$ by a numerical deconvolution. In practice, the background correction is very small when the laser is operating well above threshold, but it may become significant in the threshold region or below.

Finally, the measured counting fluctuations may require correction for the possibility that the light intensity is not constant within one counting interval. This correction becomes significant only when the counting interval T is not very small compared with the natural correlation time T_c of the light-intensity fluctuations, which we determined directly from correlation measurements. In that case the factorial variance $\langle n^{(2)}\rangle - \langle n\rangle^2$ of the photoelectric counts is no longer directly proportional to the variance of the light fluctuations, as is expected from the considerations below. Instead, we have the more general relation,[12]

$$\langle n^{(2)}\rangle - \langle n\rangle^2 = \langle n\rangle^2 \frac{2}{T}\int_0^T d\tau \left(1-\frac{\tau}{T}\right)\lambda(\tau)\,, \qquad (6)$$

in which $\lambda(\tau)$ is the normalized autocorrelation of the light-intensity fluctuations $\Delta I(t)$,

$$\lambda(\tau) \equiv \langle \Delta I(t)\Delta I(t+\tau)\rangle / \langle I\rangle^2\,. \qquad (7)$$

Evidently, if T is sufficiently short that we may replace $\lambda(\tau)$ by $\lambda(0)$ under the integral in Eq. (6), we obtain

$$(\langle n^{(2)}\rangle - \langle n\rangle^2)/\langle n\rangle^2 = \langle(\Delta I)^2\rangle/\langle I\rangle^2\,, \qquad (8)$$

which shows that the variance of the light fluctuations is directly proportional to the factorial variance of the photoelectric counts. But when this approximation is not adequate, and $\lambda(\tau)$ has the form

$$\lambda(\tau) = \sum_{r=1}^{R} C_r e^{-\lambda_r \tau}\,, \qquad (9)$$

we find from Eqs. (6) and (9)

$$\frac{\langle n^{(2)}\rangle - \langle n\rangle^2}{\langle n\rangle^2} = \frac{2}{T}\sum_{r=1}^{R}\frac{C_r}{\lambda_r}\left(1-\frac{1}{\lambda_r T}+\frac{e^{-\lambda_r T}}{\lambda_r T}\right) = \sum_{r=1}^{R} C_r\left(1-\frac{\lambda_r T}{3}+\frac{(\lambda_r T)^2}{12}-\cdots\right). \qquad (10)$$

We shall see shortly that $\lambda(\tau)$ given by Eq. (9) is indeed a reasonable approximation to the measured correlation function. Where necessary, the various corrections described have been applied to the data presented below.

III. RESULTS OF COUNTING MEASUREMENTS AND COMPARISON WITH THEORY

Because of the scaling problem that arises in making comparisons between experiment and theory, we find it more useful to concentrate on the normalized factorial moments derived from the counting measurements than on the probability distribution $p(n)$ itself. According to the Scully-Lamb[6] theory of the single-mode laser, the probability distribution $P(m)$ for the number of photons m within the laser cavity is given by the expression

$$P(m) = K_1 \prod_{r=0}^{m} \frac{A/C}{1+rB/A}\,, \qquad (11)$$

where A and C are parameters representing the gain and loss mechanisms, respectively, B is a parameter arising from the nonlinearity or saturation of the system, and K_1 is a normalizing constant. If each photon has some probability η of escaping from the cavity and registering a count at the photodetector, we may readily relate the measured counting distribution $p(n)$ to the photon distribution $P(m)$ within the cavity through a convolution with a Bernoulli distribution of parameter η, in the form[6]

$$p(n) = \sum_{m=n}^{\infty}\binom{m}{n}\eta^n(1-\eta)^{m-n}P(m)\,. \qquad (12)$$

As we show in the Appendix, the factorial variance $\langle n^{(2)}\rangle - \langle n\rangle^2$ of the number of photoelectric counts is proportional to the factorial variance $\langle m^{(2)}\rangle - \langle m\rangle^2$ of the photon number.

Generally the ratio A/B in Eq. (11) is a very large number, whereas A and C are comparable. If we put

$$A/B \equiv N_0^2\,, \quad A/C \equiv e^{a/\sqrt{2}N_0}\,, \qquad (13)$$

where N_0 is a large number and a is a positive or negative number of absolute value not very much greater than unity, we may rewrite Eq. (11) in the form

$$P(m) = K_2 N_0^{2m} e^{am/\sqrt{2}N_0}/(N_0^2+m)!\,, \qquad (14)$$

in which K_2 is another normalizing constant, and the factorial is to be understood in the sense of a Γ function. Since N_0 is a large number we use the Stirling expansion for the factorial in Eq. (14), and discard terms of order m/N_0^2 in the exponent. We then find

$$P(m) \approx K_3 \exp[-\tfrac{1}{2}(m/N_0 - a/\sqrt{2})^2]\,, \qquad (15)$$

where K_3 is another constant. If we think of the ratio m/N_0 as an almost continuous variable characteristic of the light intensity, and put

$$\sqrt{2}\, m/N_0 \equiv I\,, \qquad (16)$$

we arrive at the following probability distribution for the light intensity I:

$$\mathcal{P}(I) = \text{const} \times e^{-(I-a)^2/4}\,, \qquad (17)$$

which contains only one parameter and is precisely of the form derived from the semiclassical

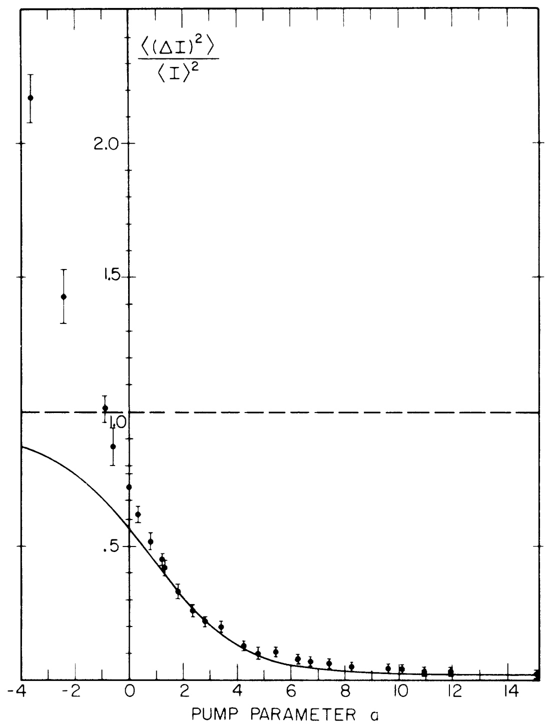

FIG. 3. Values of the relative intensity fluctuations $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ derived from counting experiments. The vertical error bars correspond to statistical uncertainty of one standard deviation. The solid curve gives the theoretical values derived from Eqs. (18) and (19).

laser theory.[5] a is the so-called pump parameter, which is positive above threshold and negative below threshold. Equation (17) leads to the following expressions[5] for the first two moments of the light intensity I:

$$\langle I\rangle = a + 2e^{-a^2/4}/\sqrt{\pi}\,[1+\mathrm{erf}(\tfrac{1}{2}a)]\,, \tag{18}$$

$$\langle(\Delta I)^2\rangle = 2 - \frac{2ae^{-a^2/4}}{\sqrt{\pi}\,[1+\mathrm{erf}(\frac{1}{2}a)]} - \frac{4e^{-a^2/2}}{\pi[1+\mathrm{erf}(\frac{1}{2}a)]^2}\,. \tag{19}$$

The mean number of counts, $\langle n\rangle$, registered in the counting measurements is, of course, proportional to the mean number of photons, $\langle m\rangle$, which in turn is proportional to $\langle I\rangle$, and, as we pointed out earlier [cf. Eq. (8)], the factorial variance is proportional to $\langle(\Delta I)^2\rangle$. In the semiclassical limit the parameter N_0 no longer enters. N_0 is actually related to the mean number of photons present at threshold through the relation $\langle m\rangle_{\text{threshold}} = N_0(2/\pi)^{1/2}$.

The results derived from the measurements for the ratio $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ are plotted against the pump parameter a in Fig. 3, for several different working points of the laser both below and above threshold. In order to relate the measurements to a and to the values derived from Eqs. (18) and (19), we needed to know the constants η and N_0 that relate photoelectric counts to photon numbers and photon numbers to the scaled light intensity I. These parameters are difficult to determine experimentally with accuracy, and we therefore resorted to curve fitting. This is a straightforward procedure when there is good agreement between theory and experiment, but is less trivial when there is disagreement. Our data indicated general agreement between theory and experiment well above threshold, and the scaling factor was therefore chosen to reflect this. However, as the curve in Fig. 3 indicates, with this choice of scaling factor there is unmistakable disagreement in the region of threshold and below, where the measured fluctuations are much larger than those predicted by Eq. (19).

In this connection, we note that in a recent theoretical treatment of the photon statistics of a dye laser, Schaefer and Willis[4] have given an expression for the probability distribution $P(m)$ of the photon number m that differs significantly from Eq. (11). Their laser model is based on four sets of energy levels for the dye molecule. There is a lower and an upper set of singlet levels, between which laser action takes place, and a lower and an upper set of triplet levels, displaced from the former, with a spacing comparable with the singlet-level spacing. Because of crossover transitions from the upper singlet states to the lower triplet states, from which molecules may be excited to the upper triplet states by absorption of laser radiation, the presence of the triplet states represents a loss mechanism for the dye laser. By incorporating the probabilities for transitions between the various levels to take place, Schaefer and Willis[4] arrived at the following expression for the probability distribution $P(m)$ of the photon number m:

$$P(m) = \text{const} \times \prod_{r=0}^{m} \frac{\nu_e NS/\nu_{Su}\nu_R}{D(r)}\,, \tag{20}$$

$$D(r) = 1 + \frac{K_{ST}}{\nu_{Su}} + \frac{2rS}{\nu_{Su}(1+2rS/\nu_{Sl})} + \frac{TK_{ST}\nu_e N/\nu_R\nu_{Su}\nu_{Tl}}{1+2T(1-\beta)r/\nu_{Tl}}\,. \tag{21}$$

Here K_{ST} is the upper singlet level to lower triplet level crossover rate, ν_{Tl} and ν_{Su} are decay rates from the lower triplet level and the upper singlet level to the lower singlet ground state, S is the stimulated emission rate from upper to lower singlet states and T is the absorption rate for transitions from lower to upper triplet states, ν_{Sl} is the fast nonradiative decay rate of the levels within the lower singlet band to the ground state, ν_e is the ground state to upper singlet excitation rate, and ν_R is the photon loss rate of the laser mode. β is the fraction of excited triplet-state molecules that return to the lower triplet states and N is the number of dye molecules that participate in laser action. According to Schaefer and Willis,[4] the ratio S/ν_{Sl} is very small, so that the term $1+2rS/\nu_{Sl}$ in Eq. (20) can be effectively replaced by unity. When the singlet-to-triplet crossover rate K_{ST} vanishes, Eq. (20) then reduces to the form of the Scully-Lamb equation (11). However, even when K_{ST} is not zero, the two equations are structurally similar when $2T(1-\beta)r/\nu_{Tl} \ll 1$, as we show below.

The values of some of the dye-laser parameters that enter into Eq. (20) have been determined by Tuccio and Strome[13] and Snavely,[14]

$$\begin{aligned} &\nu_{Su} \sim 2\times 10^8\ \text{sec}^{-1},\\ &\nu_{Tl} \sim 3\times 10^7\ \text{sec}^{-1},\\ &K_{ST} \sim 1.6\times 10^7\ \text{sec}^{-1}, \end{aligned} \tag{22}$$

although the measurements possibly refer to different solvents for the dye. These values of ν_{Tl} and K_{ST} are also consistent with cross sections determined by Peterson *et al.*[15] Schaefer and Willis[4] give the following range of values for T:

$$0.1 < T < 14\ \text{sec}^{-1}, \tag{23}$$

from which it follows that

$$2T(1-\beta)/\nu_{Tl} < 10^{-6}. \tag{24}$$

When β is very close to unity, this upper bound for $2T(1-\beta)/\nu_{Tl}$ may be further reduced by several orders of magnitude. Let us suppose that $2T(1-\beta)r/\nu_{Tl}$ in Eq. (20) is small, so that we may use the binomial expansion to write

$$\frac{1}{1+2T(1-\beta)r/\nu_{Tl}} = 1 - \frac{2T(1-\beta)r}{\nu_{Tl}} + \left(\frac{2T(1-\beta)r}{\nu_{Tl}}\right)^2 - \cdots, \tag{25}$$

and then identify the parameters as follows:

$$\begin{aligned} &\frac{\nu_e NS/\nu_{Su}\nu_R}{1+K_{ST}/\nu_{Su}+TK_{ST}\nu_e N/\nu_R\nu_{Su}\nu_{Tl}} \equiv A/C,\\ &\frac{2S/\nu_{Su}-2(1-\beta)T^2K_{ST}\nu_e N/\nu_R\nu_{Su}\nu_{Tl}^2}{1+K_{ST}/\nu_{Su}+TK_{ST}\nu_e N/\nu_R\nu_{Su}\nu_{Tl}} \equiv B/A,\\ &K_{ST}T/S\nu_{Tl} \equiv \alpha. \end{aligned} \tag{26}$$

Equation (20) then becomes expressible in the form

$$P(m) = \text{const}\times\prod_{r=0}^{m}\frac{A/C}{1+rB/A+4\alpha(1-\beta)^2(A/C)(T^2/\nu_{Tl}^2)r^2+\cdots}. \tag{27}$$

Apart from the terms in r^2, r^3, etc., whose coefficients are very small according to Eqs. (22) and (23), with

$$\alpha(1-\beta)^2(A/C)T^2/\nu_{Tl}^2 < 2\times 10^{-13}, \tag{28}$$

this equation for $P(m)$ is identical in form with the Scully-Lamb formula (11). The predictions of the Schaefer and Willis theory for the photon statistics therefore become indistinguishable from the usual simple-mode laser theory when $\alpha(1-\beta)^2(T^2/\nu_{Tl}^2)\langle m\rangle^2$ is sufficiently small and when B/A is positive. Moreover, the effect of a small contribution from these extra terms is to lower somewhat the relative second factorial moment of m [or the ratio $\langle(\Delta I)^2\rangle/\langle I\rangle^2$] near threshold. The large values of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ that we observed near threshold are therefore not accounted for by the probability distribution (20) any more than by Eq. (11).

IV. RESULTS OF CORRELATION MEASUREMENTS

In an attempt to explore the possibility that the observed excess fluctuations might not be an intrinsic feature of the dye laser, but might be connected with its mode of operation, we examined the intensity fluctuations of the pumping beam from the argon-ion laser. Figure 4 shows a photograph taken from a double-beam oscilloscope in which the top trace represents the light-intensity variation of the ion laser and the bottom trace the simultaneous variation of the dye laser. Despite the fact that the argon laser intensity fluctuates by no more than 0.2%, there is a strong indication that these small fluctuations are reflected in greatly amplified fluctuations in the output of the dye laser. However, this phenomenon does not account for all the observed excess fluctuations. Below threshold, the dye laser will even turn off at times that appear not to be correlated with downward fluctuations of the light output from the argon laser. It is possible that these effects are connected with the transit of particles in the dye solution through the pumping focus, or they may have some other origin. In any case, as Fig. 4 shows, the time scale for these phenomena appears to be significantly longer than the time scale for the natural or intrinsic laser fluctuations.

In order to explore this question further, we measured the two-time correlation function of the dye-laser intensity, as described in Sec. II. A

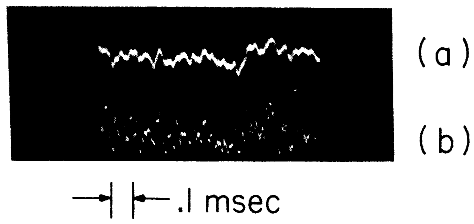

FIG. 4. Photograph taken from a double-beam oscilloscope display, showing (a) (top trace) the variation of the light intensity of the argon-ion laser, and (b) (bottom trace) the variation of the light intensity of the dye laser. The fractional intensity fluctuations are 0.002 for the argon laser and about 0.6 for the dye laser.

logarithmic plot of the number of recorded counts corresponding to the excess of $\langle I_1(t) I_2(t+\tau)\rangle$ over $\langle I_1\rangle\langle I_2\rangle$ [which is proportional to $\lambda(\tau)$] shows that there are contributions to the correlation function from two very different time constants, of which one (~7 μsec) is probably to be attributed to the intrinsic laser fluctuations, whereas the other one (~90 μsec) can probably be associated with the extraneous effects. The amplitudes of the respective contributions from these two sources can be readily extracted from the data. Figure 5 shows the result of fitting a correlation function of the form given by Eq. (9) (with $R = 2$) to the data by a least-squares procedure, from which the constants C_1, C_2, λ_1, and λ_2 can be extracted. Then the fraction of the intensity fluctuations of the dye laser attributable to intrinsic causes is given by the ratio $C_1/(C_1+C_2)$, if $1/\lambda_1$ is the shorter time constant.

By making a succession of such correlation measurements for various working points of the laser,

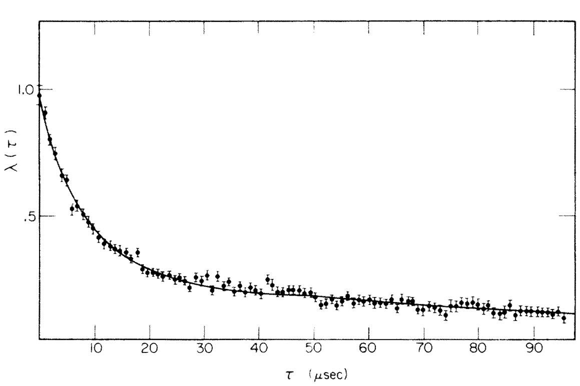

FIG. 5. Results of two-time photoelectric correlation measurements of the dye-laser intensity, superimposed on a function consisting of the sum of two exponential functions, as in Eq. (9). The function was determined by a least-squares procedure that yielded the values $C_1 = 0.68$, $C_2 = 0.30$, $\lambda_1^{-1} = 7\,\mu$sec, and $\lambda_2^{-1} = 90\,\mu$sec.

and deriving parameters by the same least-squares procedure, we were able to determine the correction factors $C_1/(C_1+C_2)$ for values of the pump parameter a below about $a = 3$. The results are given by curve B in Fig. 6. For values of a greater than about 3, the intensity fluctuations become so small that the time necessary to obtain data with sufficient statistical accuracy for curve fitting becomes prohibitively long. Fortunately, it is not too difficult to extrapolate curve B for larger values of a, where the correction factor is in any case near unity. We then used curve B to correct the values of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ shown in Fig. 3 by multiplying by $C_1/(C_1+C_2)$, so as to extract the intrinsic intensity fluctuations.

The results of this procedure are shown in Fig. 6. The extracted values of the intrinsic intensity fluctuations $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ are plotted against the pump parameter a, with the help of the relation (18) connecting a with the mean light intensity $\langle I\rangle$, and with the best choice of scaling parameter for all the data. Also shown is the theoretically predicted curve A derived from Eqs. (18) and (19) on the basis of the usual single-mode laser theory. The error bars shown correspond to a statistical uncertainty of one standard deviation, and no uncertainties associated with the correction procedure have been folded in. The correlation measurements used to correct the counting measurements were actually performed on different days, and there is some indication that the dye-laser characteristics changed gradually. Still, despite slight uncertainties in the method of correction,

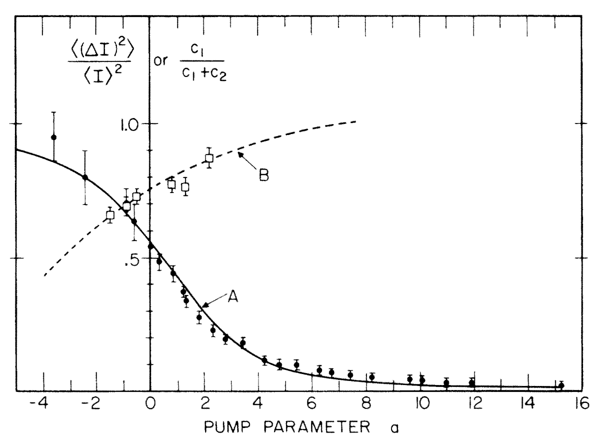

FIG. 6. Values of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ derived from the counting measurements, after correction for extraneous fluctuations as described in the text, plotted vs the pump parameter a. Curve A corresponds to the results predicted by Eq. (19) from the conventional laser theory. Curve B gives the correction factor $C_1/(C_1+C_2)$ obtained from the correlation measurements as a function of pump parameter a.

the results give no indication of any substantial departure of the intrinsic fluctuations from the conventional single-mode laser theory. Of course, this conclusion does not rule out the possibility that some of the slower fluctuations, which we labeled extraneous, may yet prove to be fundamental, although they are presently of unknown origin.

It is interesting to speculate under what conditions the extra terms in the Schaefer-Willis equation (27), corresponding to the triplet losses, might become significant and lead to changes in the observed counting statistics. As we show in the Appendix, when this happens the probability distribution $P(m)$ no longer scales in such a way that the measured moments become independent of the absolute number of photons present in the laser cavity. Since the product $\alpha(1-\beta)^2T^2/\nu_{Tl}^2$ involves the parameter ν_{Tl} to the third power in the denominator, a decrease of ν_{Tl} or a lengthening of the lower triplet-state lifetime would seem to be the obvious way to try and reveal the predicted departures. It has been suggested[4] that this might be done by reduction of the quenching agent, which, in our case, is the soap solution. However, there are technical problems to be overcome which are connected with the flow of the liquid and the effective optical pumping efficiency. We have operated our dye laser under conditions that are usual for reasonable conversion efficiency. Whether it can be operated under very different conditions in order to reveal the predicted effects is a question that remains to be explored.

ACKNOWLEDGMENTS

We are indebted to Professor C.R. Willis and Dr. R. B. Schaefer for some critical comments on an earlier version of this paper, relating to the scaling of the parameters in Eq. (20) in order to make the connection with the Scully-Lamb equation (11).

APPENDIX: RELATION BETWEEN THE FACTORIAL VARIANCE OF THE PHOTONS AND THE PHOTOELECTRONS

If each photon of the laser mode within the cavity has some probability η of escaping from the cavity and giving rise to a photoelectric pulse at the detector within some short time interval, and if these elementary processes are independent of each other, then the probability $B(n|m)$ that n photoelectric pulses will be registered when there are m photons in the cavity is given by the Bernoulli distribution

$$B(n|m)=\binom{m}{n}\eta^n(1-\eta)^{m-n}. \tag{A1}$$

If $P(m)$ is the probability that there are m photons, and $p(n)$ the probability that n photoelectric pulses are registered, then evidently

$$p(n)=\sum_{m=n}^{\infty}B(n|m)P(m). \tag{A2}$$

From Eqs. (A1) and (A2) we readily find for the factorial moments of the photoelectric counts n

$$\begin{aligned}\langle n^{(r)}\rangle &= \sum_{n=r}^{\infty}\sum_{m=n}^{\infty}n(n-1)\cdots(n-r+1)B(n|m)P(m)\\ &=\sum_{m=r}^{\infty}\sum_{n=r}^{m}\frac{m!}{(n-r)!\,(m-n)!}\eta^n(1-\eta)^{m-n}P(m)\\ &=\sum_{m=r}^{\infty}\sum_{p=0}^{m-r}m(m-1)\cdots(m-r+1)\eta^r\\ &\qquad\times\frac{(m-r)!}{p!\,(m-r-p)!}\eta^p(1-\eta)^{m-r-p}P(m)\\ &=\sum_{m=r}^{\infty}\eta^r m(m-1)\cdots(m-r+1)P(m)\\ &=\eta^r\langle m^{(r)}\rangle. \end{aligned}\tag{A3}$$

It follows as a special case that

$$\langle n\rangle=\eta\langle m\rangle \tag{A4}$$

and

$$\frac{\langle n(n-1)\rangle}{\langle n\rangle^2}=\frac{\langle m(m-1)\rangle}{\langle m\rangle^2}. \tag{A5}$$

When $\langle m\rangle$ is a large number, as is normally the case, and r is not too large, we may approximate Eq. (A3) and write

$$\langle n^{(r)}\rangle\approx\langle(\eta m)^r\rangle. \tag{A6}$$

We now show that under certain circumstances the distribution of the observed counts n may be independent of the size of the laser cavity and therefore of the mean number of photons, $\langle m\rangle$, within it. In general, when the active medium is confined to a very small region, $\langle m\rangle$ is expected to be proportional to the length of the laser cavity, and η is inversely proportional to the length. Let N_1 be a parameter proportional to the cavity length and therefore to $\langle m\rangle$. Let us write $\eta=\epsilon/N_1$, and suppose that the probability $P(m)$ of m depends only on the ratio of m to N_1, or that $P(m)$ has the form

$$P(m)=f(m/N_1)\Big/\sum_{m=0}^{\infty}f(m/N_1). \tag{A7}$$

Then it follows from Eqs. (A6) and (A7) that

$$\begin{aligned}\langle n^{(r)}\rangle &=\frac{\sum_{m=0}^{\infty}(\epsilon m/N_1)^r f(m/N_1)}{\sum_{m=0}^{\infty}f(m/N_1)}\\ &\rightarrow\frac{\int_0^\infty dy\,(\epsilon y)^r f(y)}{\int_0^\infty dy\,f(y)}.\end{aligned}\tag{A8}$$

This is independent of N_1, so that the observed counting probability $p(n)$ is independent of the length of the laser cavity, when the photon probability $P(m)$ scales as in Eq. (A7). If N_1 were proportional to N_0, then, since the Scully-Lamb formula can be reduced to the form of Eq. (15), it would satisfy the scaling condition (A7), and the same would hold true for the Schaefer-Willis formula (20) or (27) whenever the terms in r^2, r^3, etc. are very small. However, when these additional terms assert themselves, condition (A7) will not hold in general.

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*Work supported by the Air Force Office of Scientific Research and in part also by the National Science Foundation.

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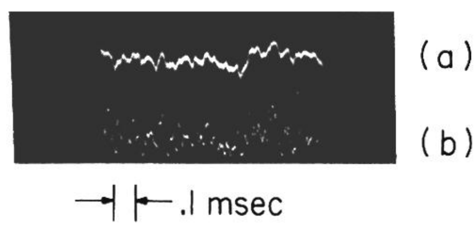

FIG. 4. Photograph taken from a double-beam oscilloscope display, showing (a) (top trace) the variation of the light intensity of the argon-ion laser, and (b) (bottom trace) the variation of the light intensity of the dye laser. The fractional intensity fluctuations are 0.002 for the argon laser and about 0.6 for the dye laser.