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Investigation of photon statistics and correlations of a dye laser

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The probability distribution $p(n)$ for the detection of n photons emitted by a single-mode dye laser in a short time at various excitations is measured, together with the distribution of time intervals between pairs of detected photons. The first measurement yields the probability density $\mathcal{P}(I)$ and the moments of the light intensity I, and the second one the two-time intensity-correlation function. Measurements are also performed at three different wavelengths. $\mathcal{P}(I)$ is found to have a two-component structure, and the relative intensity fluctuations $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ grow, apparently without limit, as the excitation tends to zero, so that no thermal state is reached. A possible explanation in terms of pumping fluctuations is discussed. The correlation function has the form of a sum of three exponential functions, and the amplitudes and decay constants of the three components are derived. The effective decay-time constant exhibits a thirteenth-power-law dependence on frequency of the laser light.

I. INTRODUCTION

It has been known since the development of the cw tunable dye laser in 1970,[1] that the emitted light has certain peculiarities as compared with more conventional lasers. For example, the dye laser generally exhibits exceptionally large intensity fluctuations when it is weakly excited. These peculiarities have sometimes been attributed to inhomogeneities in the gain medium, which is made to flow through the optical pumping region, and also to the commonly used coherent pumping mechanism, involving an auxiliary laser.

Alternatively, an entirely different and more fundamental reason for the difference may be connected with the rather complicated energy-level structure of the dye molecule, as compared with other laser systems. Besides the singlet levels involved in laser action, dye molecules usually also have excited triplet states, with an energy-level spacing that is comparable with the singlet spacing. When a nonradiative crossover transition occurs from the excited singlet state to the lower triplet state, the molecule may absorb a laser photon that raises it to the upper triplet state and causes it to act as a loss element. An excited molecule, besides exhibiting gain, may therefore also exhibit an intensity-dependent loss. The theory of such a laser has been treated by several workers,[2-4] and it has been pointed out that, because of the effect of the triplet states, a single-mode dye laser, unlike more conventional lasers, might exhibit a first-order phase transition under certain circumstances. This would be manifest in the statistics of the laser photons, which can be studied by photoelectric measurements. For example, the probability distribution of the photon number might exhibit two peaks.[2,3] If a first-order phase transition occurs, it would also be reflected in the fact that the relative fluctuations $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ of the light intensity I exceed unity near threshold, and then approach unity asymptotically from above, well below threshold.[5] Moreover, it has been demonstrated[6] that the dye laser with triplet states is mathematically equivalent to the laser with a saturable absorber, which has recently been treated in some detail.[7]

Unfortunately, the theory of the dye laser contains a large number of molecular parameters, whose values in some cases are unknown or very uncertain. This makes it difficult to know whether the predicted phenomena are actually within the range of experiment. To date no first-order phase transition has been observed in a single-mode dye laser, although such a transition has recently been seen in a ring dye laser,[8] where the effects of triplet states are less important. A few measurements of the photon statistics of a single-mode dye laser have been reported,[9-11] but the amount of quantitative information is limited. In the following we attempt to remedy this situation by measuring the probability distribution of the photon counts and the intensity-correlation function of a single-mode dye laser over a fairly wide range of excitations in the neighborhood of threshold, and at several different wavelengths. We find that the probability distribution of the light intensity exhibits a two-component structure, and that the relative intensity fluctuations become much larger than unity at very low excitations. However, this behavior seems to be connected with the on-off switching exhibited by the laser below threshold, and it is probably induced by pumping fluctuations rather than by triplet states. The wavelength dependence of the correlation times is found to be governed approximately by a 13th-power law, but this appears to be influenced at least in part by the dispersive properties of the elements of the laser.

II. EXPERIMENTAL

The dye laser is illustrated in Fig. 1. It is similar to one used in previously reported experiments,[10,11] except that the active medium is a 2×10^{-4} molar solution of rhodamine 6G dye in a 50% alcohol-water mixture. This is forced to flow continuously through a cell with quartz windows, through which the dye is optically pumped by the 5145-Å line from an argon-ion laser, as shown. This laser is stabilized in intensity at any desired power level with the help of a monitor photodiode arranged in a feedback loop. The excitation of the dye laser can be readily controlled by adjustment of the optical pumping power. The three etalons ensure single-mode operation when they are at the proper angle, and this is monitored continuously by a scanning Fabry-Perot interferometer. The three prisms shown provide the coarse dispersion. Coarse tuning of the dye laser is achieved by tilting the output mirror, and fine tuning by applying a variable voltage to the piezoelectric crystal on which it is mounted. The wavelength was varied from about 5600 to 6000 Å in these experiments. A low-gain photodetector is used to monitor the emerging light intensity, and at the higher intensities this also provides a signal for an additional control loop to keep the dye laser working-point constant. The feedback signal causes a thin glass plate mounted on the axis of a galvanometer coil, inclined a little above the Brewster angle, to rotate and this acts as a variable attenuator for fine adjustment of the

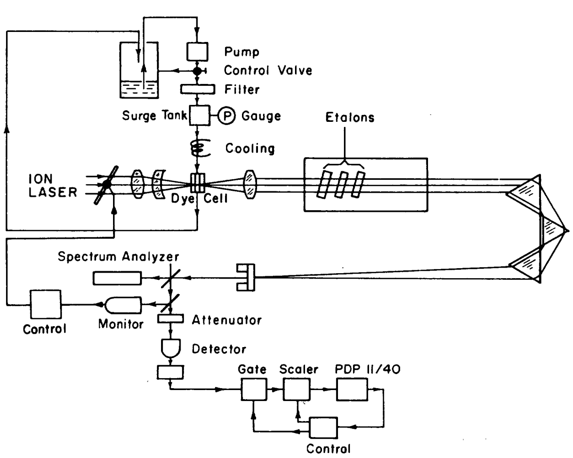

FIG. 1. Outline of the apparatus. In the correlation measurements, the output beam was divided into two by a partly silvered mirror and the two beams were allowed to fall on two photodetectors.

pumping power to the dye laser. This feedback loop is independent of the argon laser. However, no feedback was used at the lowest laser intensities. The frequency drifted no more than about 20 or 30 MHz in the course of any one measurement.

The principal measuring device for the emerging dye laser beam is an RCA 8575 photomultiplier in the photon counting measurements, as shown, and a pair of counting photomultipliers in the correlation measurements. The photoelectric pulses are passed through amplifying and pulse-shaping circuits where they are standardized, and ultimately registered by a counter. In the photon-counting measurements, the number of counts n arriving in one interval T (~1 μsec) is briefly stored in a scaler, and is then transferred to a computer memory where the information is used to increment the number stored at address n by unity. The scaler is then cleared and reset, and the system is ready for another counting cycle. After many thousands of such cycles, the number stored at address n provides a measure of the probability $p(n)$ that n photoelectric pulses are registered in a time interval T.

For values of T short compared with the intensity-correlation time of the light, $p(n)$ is simply related to the probability distribution $\mathcal{P}(I)$ of the laser light intensity I by[12]

$$p(n) = \frac{1}{n!} \int_0^\infty dI\, \mathcal{P}(I)(\alpha I T)^n e^{-\alpha I T}, \tag{1}$$

where α is a constant characteristic of the detector. If I is expressed in units of photons per second, then α is a dimensionless number that relates photoelectric counts per second to photons per second. Indeed, one can easily show from Eq. (1) that the averages are related by

$$\langle n \rangle = \alpha \langle I \rangle T, \tag{2}$$

and more generally the rth factorial moment $\langle n^{(r)} \rangle$ of n is given by

$$\langle n^{(r)} \rangle = (\alpha T)^r \langle I^r \rangle. \tag{3}$$

In particular

$$\langle n(n-1) \rangle / \langle n \rangle^2 - 1 = \langle (\Delta I)^2 \rangle / \langle I \rangle^2, \tag{4}$$

so that the relative intensity fluctuations of the light can be determined from the moments of the photoelectric counts.

In practice, corrections have to be made for background light and dark counting of the photomultiplier, by making measurements with the argon laser on, but with the dye laser extinguished. A deconvolution procedure then makes it possible to determine the probability distribution $p_L(n)$ for the laser alone from the measured distributions

$p_B(n)$ of the background alone and of $p(n)$. Alternatively, when we are focusing attention on the first two moments, we can correct the moments by writing

$$\langle n\rangle = \langle n_L\rangle + \langle n_B\rangle \tag{5}$$

and

$$\langle(\Delta n)^2\rangle = \langle(\Delta n_L)^2\rangle + \langle(\Delta n_B)^2\rangle , \tag{6}$$

where the subscripts L and B refer to the laser light alone, and the background alone. The counts also have to be corrected for the deadtime T_D(~25 nsec) of the (non-updating) counting electronics, which cause some photoelectric pulses that follow each other in rapid succession to be missed. Expressions for the first four moments of n corrected for deadtime effects have been given,[13] and a procedure for deriving the corrected probability $p(n)$ from the measured probability has been described.[14] In this work we shall mainly use the first method to correct the moments.

For the photoelectric correlation measurements, the light beam emerging from the laser is split into two beams of approximately equal intensity by a 45° beam splitter. The two beams then fall on two photomultiplier tubes whose output pulses, after amplification and pulse shaping, are fed to the start and stop inputs of a digital correlator. This device digitizes the time interval τ between start and stop pulses in steps of $\Delta\tau$(~1 μsec), so that $\tau = r\Delta\tau$, with $r = 1$ to 100, and passes the information to a computer memory.[15] After a large number N_s of counting cycles initiated by a start pulse, the number $n(\tau)$ of events with time interval τ registered becomes a measure of the two-time intensity-correlation function of the laser field. More explicitly, it can be shown that when the average counting rate in the start channel is sufficiently low,[15]

$$\langle n(\tau)\rangle = N_s R_2 \Delta\tau[1 + \lambda(\tau)] , \tag{7}$$

where R_2 is the average counting rate in the stop channel. $\lambda(\tau)$ is the normalized intensity cross-correlation function of the light intensities $I_1(t)$ and $I_2(t)$ at the start and the stop detectors, respectively, and for a stationary field it is given by

$$\lambda(\tau) \equiv \langle \Delta I_1(t)\Delta I_2(t+\tau)\rangle / \langle I_1\rangle\langle I_2\rangle . \tag{8}$$

The numbers $n(\tau)$ registered by the correlator for various delays τ therefore provide a convenient measure of the intensity-correlation function $\lambda(\tau)$.

III. RESULTS OF PHOTON COUNTING EXPERIMENTS

First several photon counting measurements were carried out with the dye laser operating at a wavelength of 5784 Å near or below threshold, where the intensity fluctuations are substantial. An oscilloscope display of the output registered by the monitor photodetector shows that the dye laser turns on and off at random times, and that the off times become progressively longer as the pumping power is reduced. A typical form of the observed probability distribution $p(n)$ is illustrated in Fig. 2. It has a high peak at $n = 0$ and a tail that extends beyond $n = 20$, but no secondary peak. From the first two moments of n we find with the help of Eq. (4) that $\langle(\Delta I)^2\rangle / \langle I\rangle^2 = 4.00 \pm 0.05$.

Unfortunately Eq. (1) is not easy to invert directly. In order to derive the probability density $\mathcal{P}(I)$ of the light intensity I from $p(n)$, we attempted to fit a function of the general form

$$\begin{aligned} (1/\alpha T)\mathcal{P}(I) = {} & (A/K)\exp(-W/K) \\ & + (B/\sqrt{2\pi}\sigma)\exp[-(W-W_1)^2/2\sigma^2] , \\ W = {} & \alpha I T . \end{aligned} \tag{9}$$

The five parameters A, B, K, W_1, σ were chosen by a least-squares procedure so that $p(n)$ derived from Eqs. (1) and (9) agreed as closely as possible with the measured values of $p(n)$. The resulting form of $\mathcal{P}(I)$ is shown in Fig. 3. It exhib-

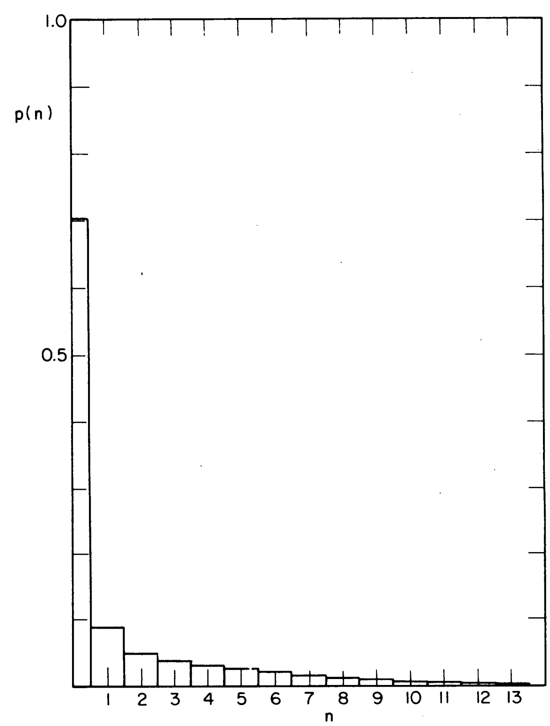

FIG. 2. Experimentally determined photon counting distribution $p(n)$, for $\langle(\Delta I)^2\rangle / \langle I\rangle^2 = 4 \pm 0.05$, at 5784 Å wavelength.

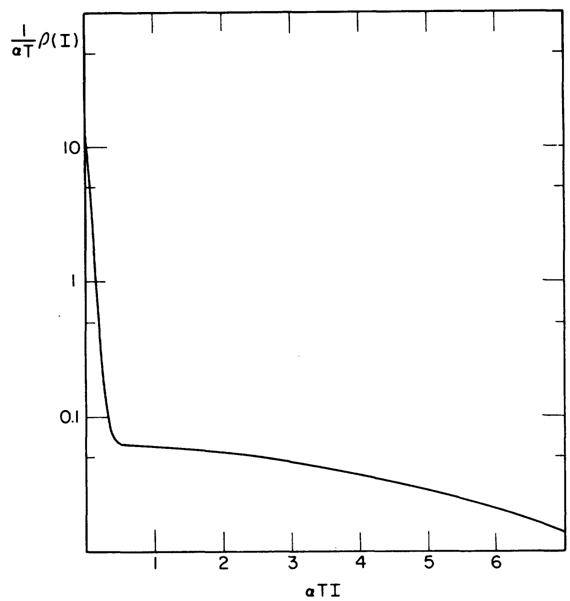

FIG. 3. Probability distribution $\mathcal{P}(I)$ of the light intensity I derived from the data in Fig. 2 with the help of Eq. (1).

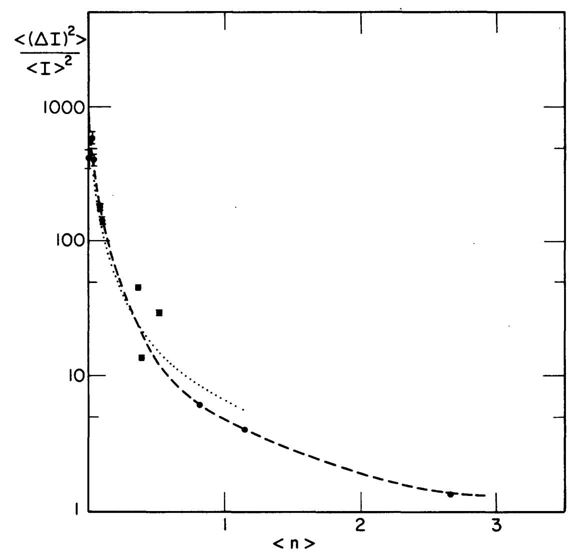

FIG. 4. Measured relative-mean-squared intensity fluctuation $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ as a function of the mean light intensity represented by the average number of counts $\langle n\rangle$. The broken curve outlines the trend. The dotted curve is derived from Eq. (17).

its a large peak at $I=0$, an almost flat region, and a long tail. Moreover, the values of $p(n)$ derived from this $\mathcal{P}(I)$ are indistinguishable from those shown in Fig. 2, so that Fig. 3 must represent a possible correct inversion of Fig. 2. The two-component form of $\mathcal{P}(I)$ bears a qualitative resemblance to some of the statistical distributions predicted for a dye laser exhibiting a first-order phase transition.[2,3] Nevertheless, the triplet states of the dye molecules are not necessarily responsible for the observed form of $\mathcal{P}(I)$, which appears to reflect the successive on-off switching of the laser; it is quite possible that the switching is caused by small-pumping fluctuations. Moreover, if a first-order phase transition did occur, this would be expected to be accompanied by hysteresis effects, which have not been observed.

As a test of the pumping fluctuation hypothesis, we have studied the intensity fluctuations over a wide range of working conditions of the dye laser, for which the average light intensity varied by more than a factor of 200. In Fig. 4 the relative variance of the light intensity derived with the help of Eq. (4) is plotted against the mean light intensity, as represented by the mean number of photoelectric counts $\langle n\rangle$ registered by the detector in 1 μsec. A few experimental points show scatter significantly greater than the standard deviation associated with the counting fluctuations, possibly because of uncompensated drift of the laser operating point. Apart from these data points the values of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ are found to increase steadily from one to several hundred as the light intensity decreases. There is no maximum of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$, as would be expected for a dye laser undergoing a first-order phase transition,[5] in which case $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ should tend to unity asymptotically from above as $\langle I\rangle\to 0$. Instead, the experimental results suggest that the laser is switched on and off at random times by optical-pumping fluctuations, and the dwell times, or on times, become progressively shorter as $\langle I\rangle$ decreases.

Let us assume for the moment that small pumping fluctuations are responsible for the observed results. If A and C are the gain and the loss rates of the dye laser,[16] then the output light intensity I above threshold $(A>C)$ may be expressed approximately in the form

$$I=G(A-C),$$

where G is a constant. The higher the excitation, the better does this equation represent the average light intensity emitted. Of course, it does not hold at all below threshold when $A<C$, and when the light intensity becomes very small. We therefore approximate the relationship between I and A by writing

$$I=G(A-C)\Theta(A-C), \tag{10}$$

where $\Theta(x)$ is the unit step function that vanishes for $x<0$. Now let the gain rate A be a randomly fluctuating variable, with Gaussian probability distribution

$$p(A) = (1/\sqrt{2\pi}\sigma)\exp[-(A-A_0)^2/2\sigma^2]\,. \tag{11}$$

Then if $A_0 < C$, the dye laser will be operating below threshold and effectively off most of the time, because the average gain A_0 does not exceed the loss. However, the laser will turn on from time to time, whenever a fluctuation causes A to exceed C. We can therefore calculate the average light intensity $\langle I\rangle$ and the variance $\langle(\Delta I)^2\rangle$ from Eq. (10) by averaging over the pumping fluctuations.

Thus we find by straightforward integration

$$\begin{aligned}\langle I\rangle &= G\int_C^\infty (A-C)p(A)dA\\ &= G\int_C^\infty [(A-A_0)-(C-A_0)]p(A)dA\\ &= G\{(\sigma/\sqrt{2\pi})\exp[-(C-A_0)^2/2\sigma^2]\\ &\qquad -\tfrac{1}{2}(C-A_0)[1-\mathrm{erf}((C-A_0)/\sqrt{2}\sigma)]\}\end{aligned} \tag{12}$$

and

$$\begin{aligned}\langle I^2\rangle &= G^2\int_C^\infty (C-A)^2 p(A)dA\\ &= G^2\int_C^\infty [(A-A_0)^2+(C-A_0)^2\\ &\qquad -2(A-A_0)(C-A_0)]p(A)dA\\ &= G^2\{\tfrac{1}{2}[\sigma^2+(C-A_0)^2][1-\mathrm{erf}((C-A_0)/\sqrt{2}\sigma)]\\ &\qquad -[\sigma(C-A_0)/\sqrt{2\pi}]\exp[-(C-A_0)^2/2\sigma^2]\}\,.\end{aligned} \tag{13}$$

If the laser is operating sufficiently below threshold that $C-A_0 \gg \sigma$, we can simplify these expressions by making use of the asymptotic form of the error function[17] for large argument,

$$1-\mathrm{erf}z \approx (1-1/2z^2+\cdots)[\exp(-z^2)]/\sqrt{\pi}z\,. \tag{14}$$

Equations (12) and (13) then reduce to

$$\langle I\rangle = [G\sigma^3/\sqrt{2\pi}(C-A_0)^2]\exp[-(C-A_0)^2/2\sigma^2]\,, \tag{15}$$

$$\langle I^2\rangle = [G^2\sigma^3/\sqrt{2\pi}(C-A_0)]\exp[-(C-A_0)^2/2\sigma^2]\,. \tag{16}$$

The inequality $C-A_0 \gg \sigma$ then makes $\langle I^2\rangle \gg \langle I\rangle^2$, so that $\langle(\Delta I)^2\rangle \approx \langle I^2\rangle$, and to a good approximation

$$\langle(\Delta I)^2\rangle/\langle I\rangle^2 = \sqrt{2\pi}Y/X\,, \tag{17}$$

where

$$\begin{aligned}Y &\equiv (C-A_0)/\sigma\,,\\ X &\equiv \sqrt{2\pi}\langle I\rangle/G\sigma\,.\end{aligned} \tag{18}$$

X is a scaled measure of the average light intensity $\langle I\rangle$. According to Eq. (15), for any given X, Y is given by the solution of the transcendental equation

$$X = (1/Y^2)\exp(-\tfrac{1}{2}Y^2)\,. \tag{19}$$

The average number $\langle n\rangle$ of photoelectric counts measured in the experiment, is of course proportional to X. If we put $\langle n\rangle = 2.36X$, we obtain the dotted curve shown in Fig. 4 from Eqs. (17)–(19). This agrees quite well with the measurements at very low intensities, and therefore lends credence to the original assumption. The same equation should not be expected to hold when $\langle n\rangle \gtrsim 1$, because the asymptotic expansion (14) is then no longer applicable, nor is the laser then operating well below threshold.

Our argument has of course been oversimplified, because we have neglected any intrinsic intensity fluctuations of the dye laser. However, as the intrinsic relative intensity fluctuations are expected to be near unity well below threshold, this neglect may not be too serious. The random modulation of the dye laser by pumping fluctuations is at least a plausible interpretation of the observed results.

IV. RESULTS OF PHOTON CORRELATION MEASUREMENTS

Two-time photoelectric correlation measurements were carried out as described in Sec. II, over a range of working points of the dye laser not too far from threshold, and at three different wavelengths. In each case the normalized intensity-correlation function $\lambda(\tau)$ was determined from the data with the help of Eq. (7). Figure 5 shows a typical set of experimental values of $\lambda(\tau)$, together with their standard deviations, obtained at a wavelength of 5627 Å.

For an electromagnetic field whose time evolution is governed by a Fokker-Planck equation, the correlation function $\lambda(\tau)$ is expected to have a structure of the form[18]

$$\lambda(\tau) = \sum_r c_r e^{-\lambda_r \tau}\,, \tag{20}$$

in which λ_r are the eigenvalues of an associated Sturm-Liouville differential equation. The coefficients c_r are positive constants expressible in terms of the eigenfunctions of the same Sturm-Liouville equation. We therefore tried to fit the experimental data by a least-squares method to an expression of this type, with three terms. The result of such a fit is shown by the full curve in Fig. 5, and it yields three coefficients c_1, c_2, and c_3 and three inverse time constants λ_1, λ_2, and λ_3. The same curve-fitting procedure was applied

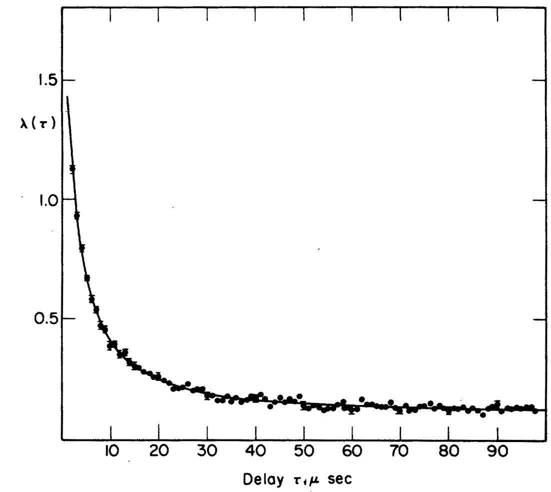

FIG. 5. Experimental values of the measured normalized intensity-correlation function $\lambda(\tau)$ as a function of τ, for $\langle(\Delta I)^2\rangle/\langle I\rangle^2 = 2.05 \pm 0.13$, and wavelength 5627 Å. The full curve is obtained by fitting the function given by Eq. (20) by a least-squares procedure.

to all the measured correlation functions $\lambda(\tau)$, and the corresponding six parameters were extracted in each case. Figures 6, 7, and 8 show the observed variation of the parameters c_1, c_2, and c_3 with the average laser-light intensity $\langle I\rangle$, at the three different wavelengths. The statistical uncertainties of the c's are rather large, so that precise functional forms of $c_r(\langle I\rangle)$ cannot be derived. Nevertheless, certain trends are clearly

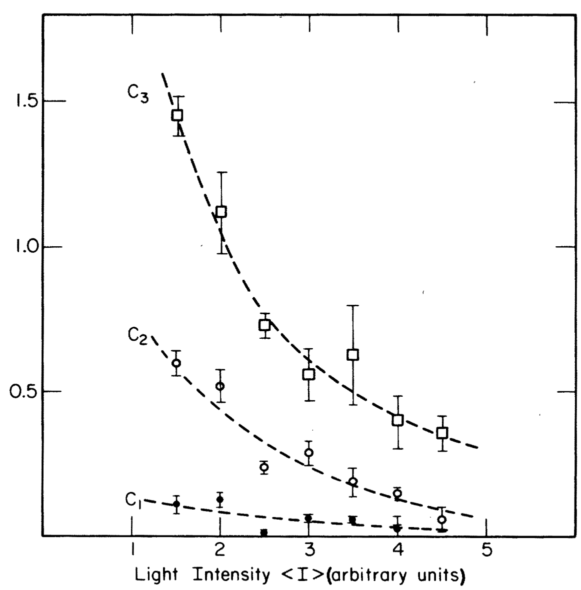

FIG. 6. Variation of the parameters c_1, c_2, and c_3 derived from the correlation measurements with average light intensity at wavelength 5627 Å.

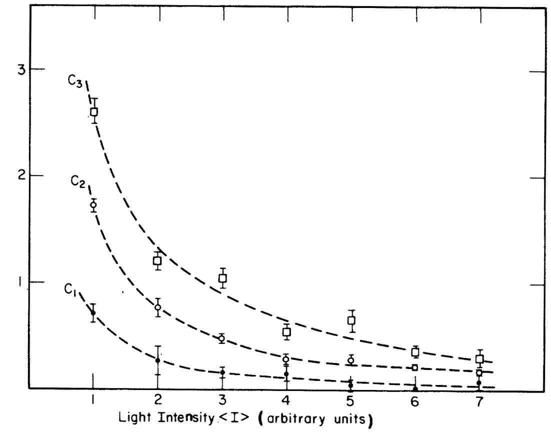

FIG. 7. Variation of the parameters c_1, c_2, and c_3 derived from the correlation measurements with average light intensity at wavelength 5784 Å.

evident. All three coefficients c_1, c_2, and c_3 rise with falling $\langle I\rangle$, and this presumably reflects the increasing magnitude of the relative intensity fluctuations as $\langle I\rangle$ decreases, that was already observed in Fig. 4. However, the relative size of the c's appears to be strongly wavelength dependent. At 5627 Å, $c_1 \ll c_2$, which in turn is less than c_3. At 5784 Å the values of the three coefficients are much closer, whereas at 5999 Å c_2 exceeds c_3, and it appears that c_1 may exceed both c_2 and c_3 at sufficiently low intensities. This implies that at low excitations the intensity variations are dominated by fast fluctuations at 5627 Å, and by slower fluctuations at 5999 Å.

From Eq. (20), the sum of the coefficients c_r

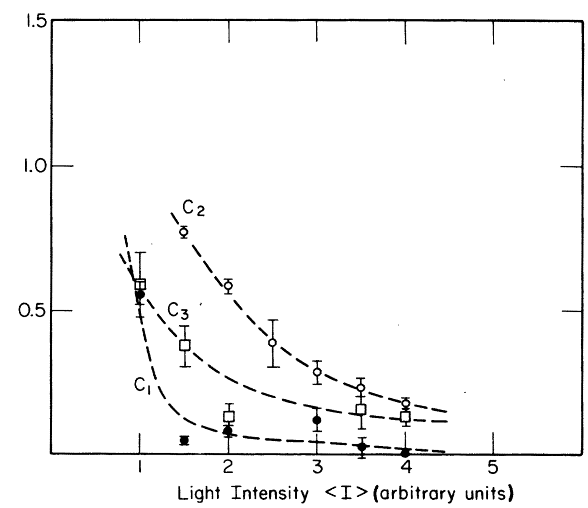

FIG. 8. Variation of the parameters c_1, c_2, and c_3 derived from the correlation measurements with average light intensity at wavelength 5999 Å.

yields $\lambda(0)$, which is also the relative intensity fluctuation $\langle(\Delta I)^2\rangle/\langle I\rangle^2$. This is plotted in Fig. 9 as a function of $\langle I\rangle$ for three different wavelengths. The range of $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ is not as large as that covered in Fig. 4, but the same trend towards increasing relative fluctuations at lower intensities is apparent. However, the scaling of the light intensity is different at different wavelengths, so that the three curves are not directly comparable.

The eigenvalues λ_1, λ_2, and λ_3 derived from the curve-fitting procedure show considerable scatter when plotted against the mean light intensity $\langle I\rangle$, and no clear trend is discernible. We have therefore averaged λ_1, λ_2, and λ_3 over the different intensities $\langle I\rangle$. However, the resulting average values are quite distinct, and, in the case of λ_2, they even exhibit a certain wavelength dependence, as illustrated in Fig. 10. However, to a first approximation, the three eigenvalues do not vary significantly with wavelength.

Nevertheless, we come to an entirely different conclusion if we construct a weighted, or effective, decay rate λ_{eff} from Eq. (20) by taking

$$\lambda_{\text{eff}} = \sum_r c_r \lambda_r \Big/ \sum_r c_r \,. \tag{21}$$

λ_{eff} is also the reciprocal time constant associated with the initial slope of the correlation function. Figure 11 shows a logarithmic plot of λ_{eff} against wavelength, from which it is quite clear that there is a substantial variation with wavelength. A straight line drawn through the points has a slope of 13, so that approximately

$$\lambda_{\text{eff}} \propto \omega^{13} , \tag{22}$$

where ω is the laser frequency.

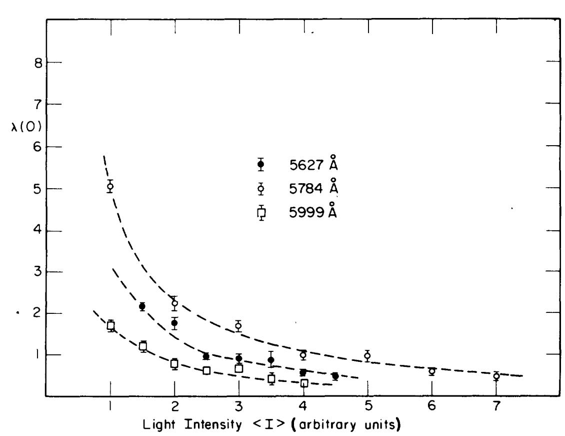

FIG. 9. Values of $\lambda(0)$ derived from the correlation measurements as a function of average light intensity $\langle I\rangle$, at three different wavelengths. The intensity calibrations are not directly comparable at the three wavelengths.

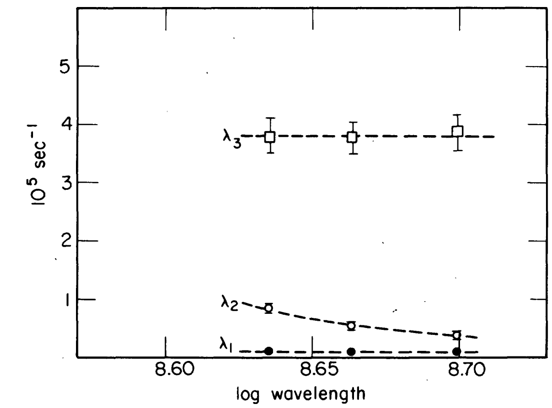

FIG. 10. Eigenvalues λ_1, λ_2, and λ_3 derived from the correlation measurements at different wavelengths.

V. DISCUSSION

Unlike the light produced by a conventional laser, such as He:Ne, the light emitted by the dye laser does not seem to reach a thermal state in which $\langle(\Delta I)^2\rangle/\langle I\rangle^2 = 1$ when the excitation is reduced sufficiently. On the contrary, so far from approaching the thermal state, the dye laser departs increasingly from it, and $\langle(\Delta I)^2\rangle/\langle I\rangle^2$ appears to increase without limit as $\langle I\rangle \to 0$. As we have seen, this behavior below threshold is probably connected with the random on-off switching induced by pumping fluctuations, as was suspected previously.[10] We have not succeeded in identifying the source of the fluctuations, but inhomogeneities and turbulence of the dye solution are possible sources.

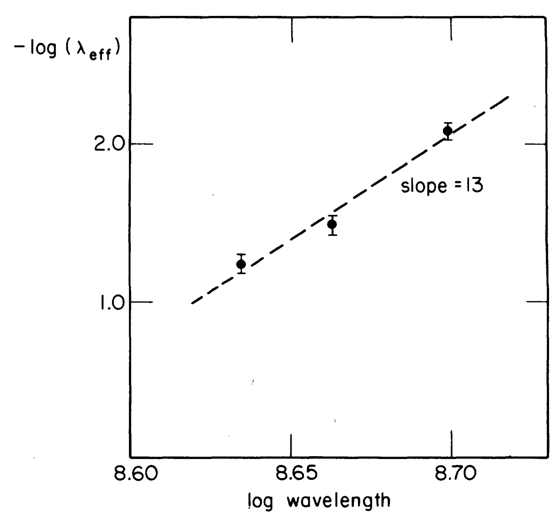

FIG. 11. Double logarithmic plot of the variation of λ_{eff} derived from the correlation measurements with wavelength.

It would of course be very desirable to be able to compare the observed correlation functions $\lambda(\tau)$ with theoretical predictions, even though there is some doubt whether all components of $\lambda(\tau)$ are associated with intrinsic—rather than possible pumping—fluctuations. Although general expressions for $\lambda(\tau)$ have been given for a laser with a saturable absorber,[3,7] which are also applicable, in principle, to the dye laser,[6] they always depend on solutions of a certain eigenvalue problem that so far have been obtained only numerically and not analytically. Some graphical solutions have been published,[3,7] but these necessarily reflect a particular choice of laser parameters, and may not apply in other cases. Unfortunately, so many of the key parameters of the dye laser are sufficiently uncertain that it does not seem practicable to make meaningful comparisons of the experimental results with the theory.

On the other hand, in principle, the frequency or wavelength dependence of the correlation times should be derivable from the conventional laser theory. It has become customary to write the equations of motion of the laser field in dimensionless scaled variables, in which time is expressed in multiples of a natural time scale for each laser. The lifetimes of the atomic states and of the field in the cavity provide such a scale. For example, from the single-mode laser theory of Scully and Lamb,[16] the natural time scale of a laser based on two-level atoms is given by $(AB)^{-1/2}$, which is also approximately equal to $(CB)^{-1/2}$ near threshold. A, C, and B are the gain, the loss, and the saturation rates of the laser, respectively. If we substitute for A, B in terms of the basic parameters of the theory, we find from the former expression that the correlation times should vary so that

$$\lambda_{\text{eff}} \propto R_2 g^3/\gamma_1^{1/2}\gamma_2^{3/2}\gamma_{12}\,, \tag{23}$$

where R_2 is the optical pumping rate, g is the coupling constant between atom and field, γ_1, γ_2 are atomic decay rates of the lower and the upper laser levels, and γ_{12} is their mean. g is generally taken to be proportional to the square root of the laser frequency ω, and if the decay rate of the upper level is governed by spontaneous emission, we would expect that $\gamma_2 \propto \omega^3$. Alternatively, if we start from the expression $(CB)^{-1/2}$, we are led to the conclusion that

$$\lambda_{\text{eff}} \propto \sqrt{C} R_2^{1/2} g^2/\gamma_1^{1/2}\gamma_2\gamma_{12}^{1/2}\,. \tag{24}$$

Of course a dye molecule is very different from a two-level atom, even when the triplet states are ignored. Still, if we assume that $\gamma_1 \gg \gamma_2$, as usual,[2,3] that the decay from the lower level is nonradiative, and that γ_1 does not depend greatly on ω, then Eq. (23) leads to the conclusion that

$$\lambda_{\text{eff}} \propto \omega^{-3}\,, \tag{25}$$

whereas from Eq. (24)

$$\lambda_{\text{eff}} \propto \sqrt{C}\omega^{-2}\,. \tag{26}$$

Both Eqs. (25) and (26) imply a frequency dependence of λ_{eff} that is rather different from, and much weaker than, the observed dependence given by Eq. (22). Even if the various optical elements of the cavity were highly dispersive, as reflected in a strong frequency dependence of the loss rate C, Eq. (26) does not adequately account for the observed results, because it would require that $C \propto \omega^{30}$. The loss rate would then be expected to increase almost sevenfold between wavelengths of 6000 Å and 5627 Å, whereas the pumping power increased only by about 20% over this range. Of course, it is conceivable that for some reason the pumping becomes much more efficient at the shorter wavelengths.

The experimental findings are therefore presented here as characteristic of one kind of dye laser, but they are probably typical of others. The results may serve as a challenge to a more detailed and more definitive theoretical treatment.

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