Correlation Functions of a Dye Laser: Comparison between Theory and Experiment

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Experimentally determined intensity correlation functions of a single-mode dye laser are compared with theoretically predicted forms given in a recent paper by Graham, Höhnerbach, and Schenzle. Although good agreement is obtained in some cases for which the parameters are chosen for best fit, large discrepancies appear for other working points of the laser. These results suggest that the dynamical theory of the dye laser needs to be modified.

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It is well known from several experiments^{1, 2} that the behavior of a single-mode dye laser in the region of threshold is not well described by the usual single-mode laser theory. Although the theory has been modified to include possible contributions from triplet states of the dye molecules,^{3,4} it does not appear that triplet states alone can adequately account for the observed switching behavior of the laser near threshold.

We have suggested² that small pumping fluctuations, possibly connected with the flow of the dye or with the ion laser that pumps the dye laser, may be responsible for the observed effects. Measurements of the relative intensity fluctuations $\langle (\Delta I)^2 \rangle / \langle I \rangle^2$ as a function of $\langle I \rangle$ were found to be reasonably consistent with such a hypothesis.² More recently a similar idea has been further developed into a dynamical theory by Graham, Höhnerbach, and Schenzle^{5,6} and Schenzle and Brand,⁷ in which the fluctuations of the pump parameter are included in the equation of motion as a form of multiplicative noise. The intrinsic quantum or spontaneous emission fluctuations, on the other hand, are assumed to be comparatively unimportant above threshold.

The theory of Graham, Höhnerbach, and Schenzle⁵ and Schenzle and Brand⁷ gives not only quite a good account of the observed instantaneous fluctuations of the laser field, but it also allows the time development of the intensity fluctuations to be tested against experiment. By choosing parameters for best fit with the data, Graham, Höhnerbach, and Schenzle succeeded in obtaining excellent agreement between one of the measured and one of the predicted two-time intensity correlation functions,⁵ which suggests that the theory is on the right track. However, one curve is not decisive, because the theory contains a free parameter; one really should look at a family of intensity correlation functions at different working points of the laser for a more searching test of the model. When such a test is carried out, quite large discrepancies between theory and experiment are encountered, which suggests that the dynamical aspects of the laser model need to be modified.

Graham, Höhnerbach, and Schenzle⁵ and Schenzle and Brand⁷ describe the laser on resonance by a dimensionless equation of motion of the usual form,

$$\mathcal{E}(t) = \left[a_1(t) - A_1 \right] \mathcal{E}(t) \left[{}^2 \right] \mathcal{E}(t) , \qquad (1)$$

in which $\mathcal{E}(t)$ is the single-mode laser field, and A_1 is a constant describing the saturation. However, $a_1(t)$ is taken to be a fluctuating pump parameter, which is regarded as a δ correlated, Gaussian random process of mean a_{10} ,

$$\langle [a_1(t) - a_{10}][a_1(t') - a_{10}] \rangle = Q_{11} \delta(t - t').$$
 (2)

Equation (1) has been solved,^{6,7} and the steadystate distribution $W_0(I)$ of the light intensity $I \equiv |\mathcal{E}|^2$ above threshold $a_{10} > 0$ takes the form of a γ distribution,⁵

$$W_0(I) = \left[q^{\alpha} I^{\alpha-1} / \Gamma(\alpha) \right] \exp(-qI), \qquad (3)$$

with

$$q \equiv Q_{11}/4A_1, \quad \alpha \equiv a_{10}/Q_{11}.$$
 (4)

It follows from this that the relative intensity fluctuations are given by

$$\lambda(0) \equiv \langle (\Delta I)^2 \rangle / \langle I \rangle^2 = 1/\alpha , \qquad (5)$$

and the mean intensity is

$$\langle I \rangle = \alpha/q = 4A_1 \alpha/Q_{11}, \qquad (6)$$

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so that

$$\lambda(0) = 4A_1 / Q_{11} \langle I \rangle. \tag{7}$$

If the noise parameter Q_{11} is taken to be independent of the working point of the laser, then $\lambda(0) \propto 1/\langle I \rangle$.

$$\lambda(\tau) = \frac{\exp(-\alpha^2 \tau)}{16\alpha^2 \Gamma(\alpha)} \int_{-\infty}^{\infty} dx \, \frac{(\alpha^2 + x^2)x \sinh(\pi x) \left| \Gamma(\frac{1}{2}\alpha + \frac{1}{2}ix) \right|^2 \exp(-x^2 \tau)}{\cosh(\pi x) - \cos(\pi \alpha)} , \tag{9}$$

with $\tau \equiv \frac{1}{2}Q_{11}t$. The two parameters α and Q_{11} therefore completely determine the form of the predicted correlation function $\lambda(t)$.

We start by testing for the predicted reciprocal relationship between $\lambda(0)$ and $\langle I \rangle$ when Q_{11} is constant. Figure 1 shows measured values² of $\lambda(0)$ at various laser intensities $\langle I \rangle$ for three different wavelengths, superimposed on theoretical curves of the form given by Eq. (7). Although the experimental points exhibit a fair amount of scatter, the data are generally consistent with the hypothesis that Q_{11} is independent of the laser working point at any one wavelength. If Q_{11} should vary with $\langle I \rangle$, it cannot be by more than 10% or 15% over the range in question, and certainly not by a factor 3 or 4.

Let us now compare the measured and predicted



The solution of the time-dependent equation leads to the following integral expression for the normalized intensity correlation function⁵⁻⁸

$$\lambda(t') \equiv \langle \Delta I(t) \Delta I(t+t') \rangle / \langle I \rangle^2 \tag{8}$$

for the case $\alpha < 2$:

forms of $\lambda(t)$ when Q_{11} is constant. The correlation functions $\lambda(t)$ measured in Ref. 2 were all found to be well described by the sum of three exponentials, at least one of which has a rather long time constant.² Graham, Höhnerbach, and Schenzle have argued that their theory should not be expected to account for the long tail, which should be subtracted out.⁵ They have therefore fitted the theory not to the experimentally determined correlation function² $\lambda(t)$, but rather to $\lambda(t) = \lambda(100 \ \mu sec)$ in the range $t < 100 \ \mu sec$ only. For consistency we have followed the same method below.

Our procedure, which is similar to theirs, is to start from the measured values of $\lambda(t) - \lambda(100)$ μ sec), as given in Ref. 2, which yields α according to Eq. (5) with an accuracy of order 10% or better. With this value of α Eq. (9) is then used to derive the parameter Q_{11} that produces the best fit with the corresponding correlation $\lambda(t)$.



FIG. 1. Comparison of the experimental and theoretical relationship between $\lambda(0)$ and $\langle I \rangle$ at three different wavelengths. The broken curves are based on Eq. (7).

FIG. 2. Comparison of theoretical and experimental correlation functions at 5627 Å, for the case $\alpha = 1/\lambda(0)$ = 0.52. The full curve is experimental from Ref. 2, and the broken curve is derived (Refs. 5 and 8) from Eq. (9) with $Q_{11} = 0.18 \ \mu \text{sec}^{-1}$.



FIG. 3. Comparison of theoretical and experimental correlation functions at 5627 Å, for the case $\alpha = 1/\lambda(0)$ = 1.05. The full curve is experimental from Ref. 2, and the broken and dotted curves are derived (Refs. 5 and 8) from Eq. (9).

With the help of our data for $\alpha = \frac{1}{2}$, Graham, Höhnerbach, and Schenzle⁵ have concluded that $Q_{11} = 0.18 \ \mu \text{sec}^{-1}$. We use the same value of Q_{11} below, together with the different derived values of α , to compare the theoretical forms of $\lambda(t)$ given by Eq. (9) with the measured forms,² for different working points of the laser.

The results of such a comparison are shown in Figs. 2-4. In each case the full curves are experimental, and they are extracted from the data by a least-squares procedure. Figure 5 of Ref. 2 gives an indication of the dispersions of the actual data points about the least-squares curve, which is quite well defined. The broken curves are theoretical and they are derived from Eq. (9) with Q_{11} and α obtained as described above. In Fig. 2 the agreement is excellent, but this is the data set for which Q_{11} was chosen for best fit. It corresponds to Fig. 3 of Ref. 5. For other working points of the laser one finds progressively worse fit, and by the time $\alpha \ge 1$, the disagreement between theory and experiment is guite substantial. The situation would be somewhat worse even, if the subtractions described above were not made.

It is possible to improve the agreement by adjusting Q_{11} each time for best fit with the data, but this requires large changes of Q_{11} . The dotted curves in Figs. 3 and 4 are obtained when Q_{11} is adjusted in this way and treated as a free parameter each time. Although the determination



FIG. 4. Comparison of theoretical and experimental correlation functions at 5627 Å, for the case $\alpha = 1/\lambda(0)$ = 1.84. The full curve is experimental from Ref. 2, and the broken and dotted curves are derived (Refs. 5 and 8) from Eq. (9).

of the best-fitting curve is somewhat subjective, it is clear from Figs. 3 and 4 that even the best one does not always fit very well. Moreover, the values of Q_{11} required for best fit in Figs. 3 and 4 differ so substantially (by more than a factor of 3) from the Q_{11} value derived earlier from Fig. 2 as to contradict completely the inverse proportionality between $\lambda(0)$ and $\langle I \rangle$ that is clearly demonstrated by the data in Fig. 1.

We are therefore led to conclude that the correlation properties of the dye laser are not always well described by Eq. (9). It is possible that the fluctuations of the pump parameter are not sufficiently rapid to be representable by a δ correlated noise. Indeed, there is some evidence that the pumping fluctuations may be slower, rather than faster, than the intrinsic intensity fluctuations of the laser field.¹ Although the idea of Graham, Höhnerbach, and Schenzle⁵ and Schenzle and Brand⁷ that the dye laser behavior is dominated by pumping fluctuations may yet turn out to be correct, the challenge to construct an adequate dynamical theory remains.

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Structure of Wall Plasmas near Divertor Neutralizer Plates or Limiters

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The structure of wall plasmas near neutralizer plates or limiters is calculated with a kinetic transport model which includes binary collision processes between the various ion, atomic, and molecular species, and the self-consistent electrostatic field. A family of solutions is found which may be used in calculating removal of heat and impurities.

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This Letter reports a self-consistent solution to the problem of the structure of a plasma adjacent to a solid surface, such as a divertor neutralizer plate or limiter. A plasma near such a surface will be affected by the cold gas collisionally released from the surface. In steady state the return flux of cold gas replaces the ion flux burying itself in the surface, and contributes, in its various ionization states, a significant component to the plasma structure. This structure, with its accompanying electrostatic field, controls heat and impurity transport to the surface, and sputtering from the surface. Understanding its nature is fundamental to the general question of plasma confinement, and particularly to the operation of divertors and limiters.

For plasma densities and temperatures envisioned in divertor chambers the mean free paths for momentum exchange are on the order of, or long relative to, mean ionization lengths for a cold species in the plasma. Near a surface the plasma thus will be far from local thermodynamic equilibrium (LTE), and the electrostatic potential will strongly influence the distribution functions of the slow ions.¹ To specify the potential, however, requires knowledge of these distribution functions. The problem thus demands an approach which will generate both distribution functions and potential self-consistently.

Presented here are the results of such a calculation for a Maxwellian hydrogen plasma with ion temperature T_i incident on a surface which is reemitting molecular hydrogen H₂⁰ at a surface temperature T_s . The calculation is kinetic in the sense that the ions and electrons are subject to acceleration by the electric field only; momentumtransfer collisions are ignored. This is a reasonable assumption for T > 50 eV. The calculation assumes a steady state, and is limited to an (x, v_x) phase space, where x is normal to the surface. Electrons are taken to have a spatially isothermal Boltzmann distribution with a temperature T_e equal to the incoming ion temperature, T_i . The ion flux to the surface is assumed to be completely absorbed, and the return molecular flux is some multiple R (by mass) of this incident flux.

Collisions with ions and electrons will ionize and dissociate the cold gas flowing from the surface, and are included in the calculation. The