Colored-noise-induced first-order phase transition in a single-mode dye laser

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We report the observation of a colored-noise-induced first-order nonequilibrium phase transition in a single-mode dye laser. Although a second-order phase transition exists in the more usual single-mode laser models, the order of the phase transition can be altered by including colored-noise fluctuations in the pumping of the laser. The nonwhite character of the noise is shown to be crucial to the onset of the characteristics of a first-order phase transition.

INTRODUCTION

The single-mode laser has been a useful physical system in which to study noise in nonlinear dynamical systems. Models including both additive white and either white or colored multiplicative noise have been used to model the photon statistics of a dye laser.[1] Here we show that the single-mode dye laser can undergo behavior analogous to a first-order phase transition. The discontinuous change of the most probable intensity as a function of pumping shows this analogy. The transition is induced by fluctuations in the effective pumping of the system, and we stress that it is the nonwhite character of these fluctuations that causes the transition to be of the first-order type instead of the second order.

Lasers have long been a primary resource for both experimental and theoretical investigations of nonequilibrium phase transition analogies. In this vein, the single-mode laser threshold has long been known to display a second-order phase-transition analogy.[2] Indeed, this discovery marked the real beginning of the study of nonequilibrium phase transitions. Variations on the phase-transition analogy in the single-mode laser have been studied also. In particular, phase-transition analogies of the first order have been sought. The single-mode laser with saturable absorber[3,4] is known to exhibit such an analogy. Multimode laser systems have also been examined in this light and a first-order phase transition has been predicted and observed in the two-mode dye laser.[5] Recently there has been speculation about the cause of yet another first-order phase-transition analogy in multimode dye lasers.[6] In that case the phase transition concerns discontinuous jumps in the power spectral density of the multimode dye laser.

On a related front, the study of noise-induced transitions has also been fruitful and, in particular, the study of noise in nonlinear dynamical systems has produced interesting results.[7] The study of nonwhite noise in these systems has stimulated even more interest. Colored-noise-induced transitions have been studied by Kitahara, Horsthemke, and Lefever.[8] These studies stress the robustness of the noise-induced transitions under the change from a white-noise to a "colored"-noise driving term. Here we emphasize that the nonzero correlation time can have its own consequences. In this example the nature of the phase transition is changed from the second order to the first order.

The study of noise in lasers, which are relatively simple nonlinear dynamical systems, has been particularly active within the last few years. This study, concentrating on the modeling of the intensity correlations in dye lasers with noisy pumps, has been quite productive.[1] The consensus that has emerged from these studies is that the best model of the dye laser is that of the "standard" stochastic model[9] with a colored-noise fluctuating pump added:

$$\dot{E}=(a-|E|^2)E+\eta(t)E+q(t)\ , \tag{1}$$

$$\langle q^*(t)q(t')\rangle=4\delta(t-t'),\ \ \langle q(t)\rangle=0 \tag{2}$$

$$\langle \eta^*(t)\eta(t')\rangle=Q\Gamma e^{-\Gamma|t-t'|},\ \ \langle \eta(t)\rangle=0\ . \tag{3}$$

Here E is the complex field amplitude, a is the pump parameter, and $q(t)$ and $\eta(t)$ are complex stochastic noise terms representing spontaneous emission into the system and fluctuations in the pumping, respectively. In this dimensionless form $q(t)$ is scaled to have a fixed noise strength and the colored noise η is an Ornstein-Uhlenbeck process with a strength Q and a bandwidth Γ.

Here we bring together all of these topics and put forth evidence for a colored-noise-induced phase transition in the single-mode dye laser with a noisy pump. Although evidence allowing the prediction of this transition has appeared in the literature,[10,11] the analogy appears not to have been pointed out before. Evidence of its existence in an experimental system is also presented.

THEORY

The fact that the system of equations (1) and (2) with white-noise pump fluctuations,

$$\langle \eta^*(t)\eta(t')\rangle=2Q\delta(t-t')\ , \tag{4}$$

does not show the equivalent phase transition has probably obscured the evidence for the transition in the past.

If we examine a simpler model, without including spontaneous emission fluctuations, we can show that this first-order phase transition is indeed colored-noise induced. This was already evident in the work of Sancho *et* *al.*[10] To this end, we first examine the one-dimensional white-noise model for the field amplitude:

$$\dot{x} = ax - x^3 + \eta(t)x \ , \tag{5}$$

$$\langle \eta(t)\eta(t') \rangle = 2Q\delta(t-t'), \ \ \langle \eta(t) \rangle = 0 \tag{6}$$

whose solution in the steady state is[12]

$$P(x) = N' x^{-1+a/Q} e^{-x^2/2Q} \ . \tag{7}$$

Here the intensity of the laser $I = x^2$ and

$$P_{\text{white}}(I) = N I^{a/2Q-1} e^{-I/2Q} \ , \tag{8}$$

where N and N' are normalization constants. The most probable intensity, given the distribution (8), is

$$I_{\text{MP}} = \begin{cases} 0 & \text{for } a \le 2Q \\ a - 2Q & \text{for } a \ge 2Q \ . \end{cases} \tag{9}$$

The most probable intensity is then a continuous function of a, according to (7), although its first derivative is discontinuous at the point where $a = 2Q$. This is, as in the case of Ref. 2, analogous to a second-order phase transition.

We now allow $\eta(t)$ to be a (real) colored noise, modeled by an Ornstein-Uhlenbeck process,

$$\langle \eta(t)\eta(t') \rangle = Q\Gamma e^{-\Gamma|t-t'|}, \ \ \langle \eta(t) \rangle = 0 \ . \tag{10}$$

We must now combine Eq. (5) with the following relations,

$$\dot{\eta} = -\Gamma\eta + \Gamma\sqrt{Q} f(t) \tag{11}$$

$$\langle f(t)f(t') \rangle = 2\delta(t-t'), \ \ \langle f(t) \rangle = 0 \tag{12}$$

to complete the model. In this case the only analytic solution to the two-dimensional problem available is the matrix continued-fraction solution of Jung and Risken[11] which requires numerical evaluation to determine its structure. They have done this and produced distributions for $P(I)$ which are given in Ref. 11. These plots demonstrate a two-peaked distribution in I and hint at a first-order phase transition in I_{MP} as a function of either Q or Γ.[13] Thus, this evidence is suggestive of a first-order phase transition with a being the control parameter.

Sancho *et al.* have worked on an approximate solution to this problem, and the first evidence of this transition is given in Ref. 10. They derive an equation for x in a large-Γ approximation. In this approximation the solution to (5), subject to Eq. (10)—(12), is

$$P(I) = P_{\text{white}}(I) \left[1 - \frac{1}{\Gamma} \left[\frac{a^2}{2Q} + 1 - \frac{2Q+a}{Q} I + \frac{I^2}{2Q} \right] \right] , \tag{13}$$

where P_{white} was given in (8).

Although Eq. (13) is not particularly transparent in form, Sancho *et al.* have performed an *ad hoc* exponentiation of this result in order to extend their result to smaller Γ and, in the process, they have created a more tractable expression. Considering the term in square brackets in (13) to be the first two terms of a Taylor expansion, they write

$$P(I) = N I^{a/2Q-1} \exp \left[-\frac{a}{\Gamma} \frac{2Q+a}{2Q} + \left[\frac{4Q+2a-\Gamma}{2Q\Gamma} \right] I - \frac{I^2}{2Q\Gamma} \right] . \tag{14}$$

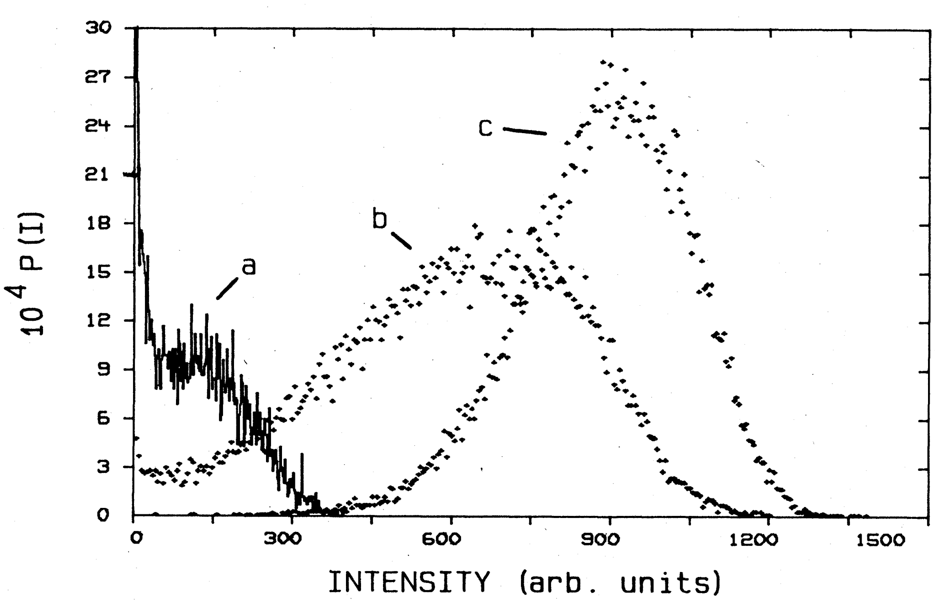

FIG. 1. Normalized distributions of the experimental laser intensity $P(I)$ taken at three different laser working points. Curve a, $\langle I \rangle = 32$. The initial point of the distribution, $P(0)$, is approximately 50. Curve b, $\langle I \rangle = 590$. The initial point of the distribution, $P(0)$, is approximately 0.54. Curve c, $\langle I \rangle = 890$, $P(0) \approx 0$. Intensities are given in arbitrary units.

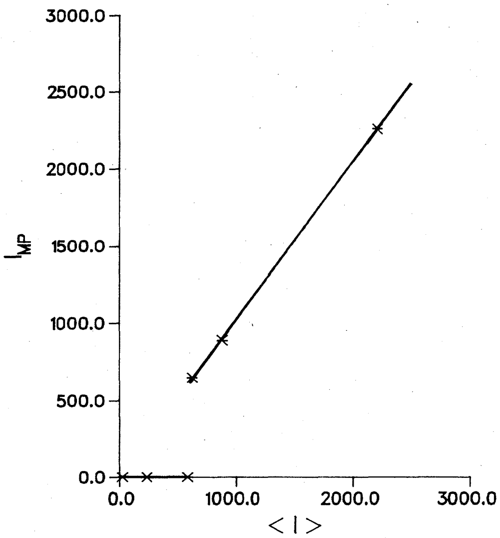

FIG. 2. Plot of the experimentally determined most probable intensity vs the mean intensity of the dye laser showing the first-order phase-transition analogy.

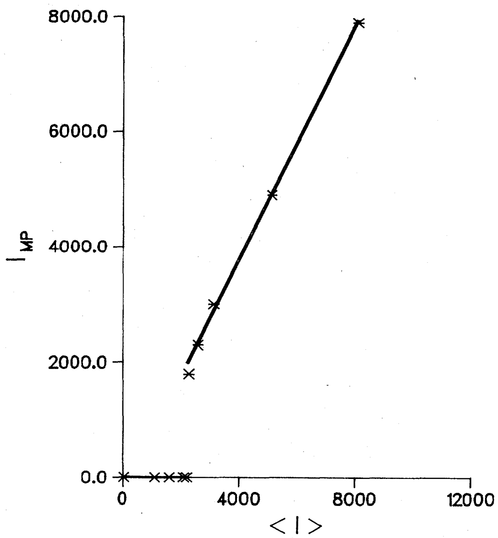

FIG. 4. Plot of the most probable intensity vs the mean intensity taken from simulations of Eqs. (1)–(3). The noise parameters were fixed at $Q=5000$ and $\Gamma=1000$.

The extrema of (14) are given by

$$\frac{2}{\Gamma}\frac{I^2}{a}-\left[2\left[\frac{2Q}{a}+1\right]\frac{a}{\Gamma}-1\right]\frac{I}{a}+\frac{2Q}{a}-1=0 \quad \text{with } I\geq 0 . \qquad (15)$$

For $a>2Q$ (15) gives a unique positive value for the maximum of the distribution (14). For $a<2Q$, however, $I=0$ is a local maximum and, if Γ is small enough, another local maximum and a minimum appear in the distribution. The positive roots of (15) are discussed by these authors and the regions of one and two maxima of $P(I)$ are outlined in that reference. A "phase diagram" of this system is presented there also. Although it is not shown there that the system can be continuously driven through what would be called a first-order phase transition, it is obvious now that this can occur.

EXPERIMENTAL PROCEDURE

The dye laser used for these measurements has been described previously.[14] It consists of a three-mirror standing-wave cavity with three intracavity etalons to maintain single-mode operation at all pumping levels.

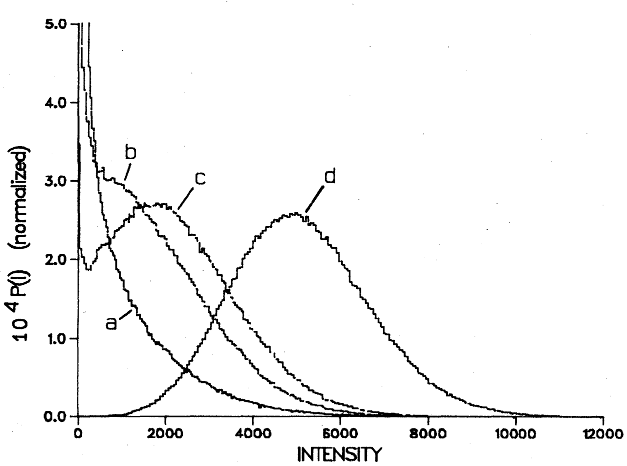

FIG. 3. Distributions of laser intensity $P(I)$ as extracted from simulations of Eqs. (1)–(3), taken with four different average pump parameters: curve a, $a=400$, curve b, $a=1000$, curve c, $a=2100$, and curve d, $a=5000$. The noise parameters were fixed at $Q=5000$ and $\Gamma=1000$.

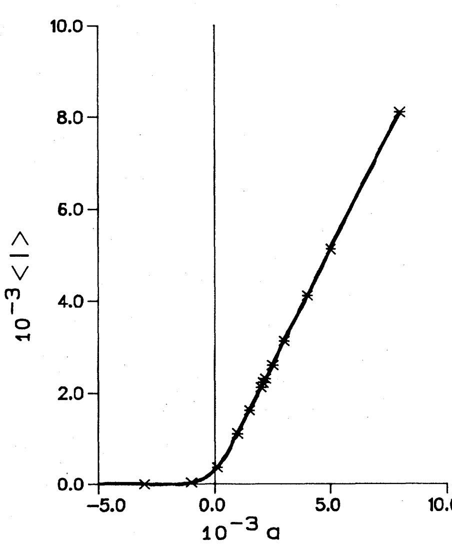

FIG. 5. Plot of the mean intensity vs the true control parameter, the pump parameter a. The noise parameters were fixed at $Q=5000$ and $\Gamma=1000$.

Pumping of the dye laser was achieved by focusing an argon-ion laser into the flowing gain medium in a dye cell with quartz windows. The dye solution was 2×10^{-4} molar rhodamine 6-G in an alcohol and water solution. The tight focusing of the pump laser, combined with any turbulence in the dye flow, can cause rather large fluctuations in the effective pumping of the system. The light output was gathered and focused onto a fast photodiode. The voltage output was amplified and fed into a fast analog-to-digital converter which was interfaced to a computer. Approximately 30 000 data points were collected at 10—200 kilosamples per second. These samples had 11 bits of resolution and were sorted into 250 bins for display. The mean output intensity of the laser is determined independently with a second, integrating, photodiode so that the gain into the transient digitizer may be adjusted. The intensity data is displayed using an arbitrary linear scale where $\langle I\rangle=2000$ corresponds approximately to 1 mW of power at ~6000 Å.

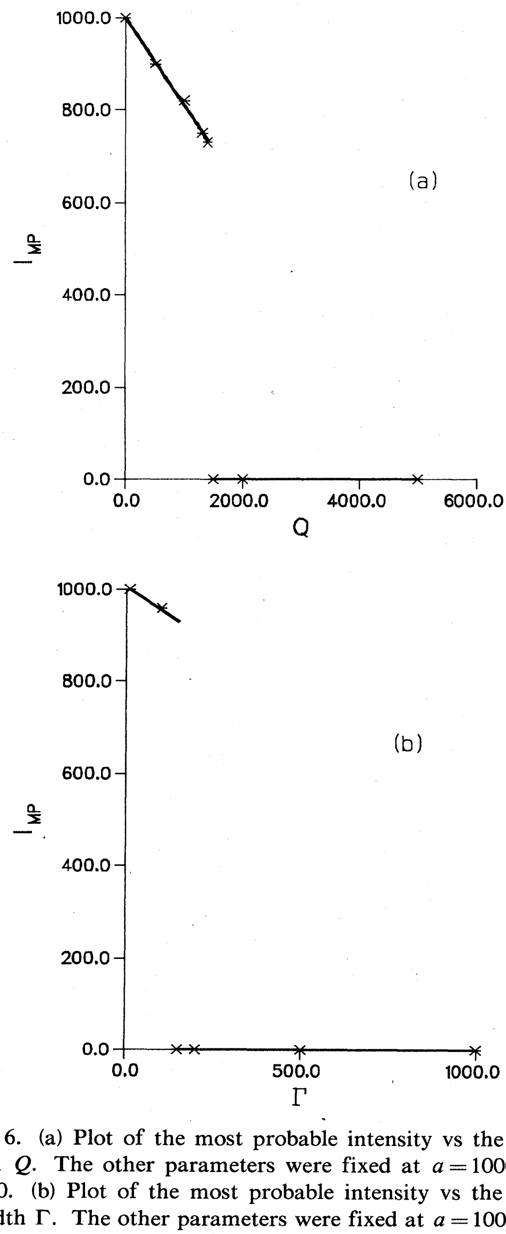

FIG. 6. (a) Plot of the most probable intensity vs the noise strength Q. The other parameters were fixed at $a=1000$ and $\Gamma=1000$. (b) Plot of the most probable intensity vs the noise bandwidth Γ. The other parameters were fixed at $a=1000$ and $Q=5000$.

Sample $P(I)$ distributions are shown in Fig. 1. The distributions shown indicate the behavior at three working points of the laser. At the lowest operating points the intensity distribution has a single peak at zero intensity (see Fig. 1, curve a). As the pumping is increased, the peak at zero intensity decreases while a second peak appears at a nonzero value of the intensity (see Fig. 1, curve b). At some point this second peak becomes larger than the zero-intensity peak. At working points well above threshold we find only a single peak in the distribution at a nonzero intensity value (see Fig. 1, curve c).

If we plot the value of the most probable intensity, I_{MP}, versus the mean value of the intensity, $\langle I\rangle$, we see a standard first-order phase-transition analogy. This is shown in Fig. 2. Well above threshold $\langle I\rangle$ is a linear function of the pump parameter while near threshold it is rather slowly varying with the pump strength. Although it might be preferable to plot I_{MP} versus the pump parameter a, this parameter is impossible to determine from the model (5) with (11) and (12) unless one knows the parameters Q and Γ.[14,15]

Monte Carlo simulations were performed using the model (1)—(3). These simulations are described elsewhere.[14,15] Representative results of these calculations are shown in Fig. 3. It is again obvious that there is a discontinuous jump in the most probable intensity as the pump parameter is varied. The most probable intensity, as a function of the mean intensity, is plotted in Fig. 4. The parameters Q and Γ are given definite values in order to perform the simulations. Plotting versus pump parameter is, in this case, possible. The relationship of the true control parameter a (approximately a linear function of the pump intensity) to the mean intensity $\langle I\rangle$ is given in Fig. 5.

As suggested by the work of Sancho *et al.*[10] the phase transition is also visible as a function of both the noise strength Q and the noise bandwidth Γ. The first-order phase transition in I_{MP} for the model (1)—(3) is shown in Figs. 6(a) and 6(b) as a function of Q and Γ, respectively.

DISCUSSION

First-order phase transitions were predicted in models of dye lasers that included triplet states some years ago by Schaefer and Willis[16] and Dembinski and Kossakowski.[17] Roy[18] has shown that the singlet-triplet model used by these authors is completely equivalent to the laser with a saturable absorber. In this model the triplet states act as a built-in saturable absorber in the system. Roy and Mandel[19] have shown that, when the triplet states are allowed to play a role, large relative intensity fluctuations are to be expected. This phenomenon was already observed in earlier experiments on a single-mode dye laser,[20] but it was attributed to pumping fluctuations even at that time. Further experiments on single-mode dye lasers attempted to extract $P(I)$ by inverting the measured photocount distribution $p(n)$.[21] The inversion of $p(n)$ to obtain $P(I)$ is nonunique and the inversion technique used in that work may not have shown a double-peaked distribution for $P(I)$ even if it was present. In any event, Kaminishi *et al.*[21] concluded that there was no evidence for a first-order phase transition or for triplet effects in the single-mode dye laser.

In agreement with Kaminishi *et al.*, we explain our results here as being due to pumping fluctuations, however, we do see clear evidence of a first-order phase transition in the direct measurements of $P(I)$. The transition seen here does not seem to have its origin in the inclusion of triplet states in the dye model or the presence of a saturable absorber. Rather, its most natural explanation seems to be that it is a noise-induced transition, and can be explained in terms of a simple, phenomenological, single-mode laser model that includes pumping fluctuations. It is in a unique class, even in terms of noise-induced transitions, in that it requires not just the *presence* of a multiplicative noise but the presence of a *colored* multiplicative noise.

It should be noted that the choice of parameters in the simulations is somewhat less than certain. The simulation results presented in Figs. 3–6 are meant to verify that the model of Eqs. (1)–(3) does indeed reproduce the important phase-transition-like behavior of the dye laser. The actual values of the parameters used are not necessarily optimum, and thus the shape of the experimental distributions should not be expected to be reproduced in great detail. One feature of the fit to the experimental distributions that will not be improved by changing the parameter values is the asymmetry of the high-intensity peak. The model of Eqs. (1)–(3) seems to be unable to produce an asymmetric peak for the "on" state of the dye laser. A similar situation is found in the case of the two-mode dye laser which also exhibits a first-order phase transition.[5] Measurements of $P(I)$ for one of the two modes have been carried out[22] and an asymmetry in the "on" state peak for that mode was found. The asymmetry of the measurements in Fig. 1, curves b and c, is very reminiscent of what was seen there. At that time, the asymmetry in the two-mode laser was thought to be due to backscattering coupling the modes in the ring cavity. It is apparent, however, that unless backscattering from elements external to the cavity are affecting the laser, the single-mode results cannot be explained in the same way.

It is quite possible that the true source of this asymmetry is due to something rather more fundamental. The model (1) is a simple third-order theory of the laser and it is quite possible that higher-order saturation effects are suppressing the high-intensity side of this peak in the $P(I)$ distribution. It is easily seen that when the theory is extended to include terms of the fifth order in E, there is an asymmetry induced in the intensity distribution that resembles that seen in Fig. 1, curves b and c.

In conclusion, we point out that the single-mode dye laser with a noisy pump is another example of a system displaying a noise-induced phase-transition analogy. It is, however, the order of the phase transition which it exhibits that depends on the noise. In this case the order of the phase transition depends critically on the correlation time of the driving fluctuations in its pump.

Note added in proof. In J. M. Sancho, M. San Miguel, H. Yamazaki, and T. Kawakubo, Physica **116A**, 560 (1982), the "phases" of the system modeled by Eqs. (5) and (10) are further discussed and investigated experimentally in an electrical circuit. We thank Dr. J. M. Sancho for bringing this work to our attention.

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