# Mode-correlation times and dynamical instabilities in a multimode cw dye laser

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Correlation times of mode amplitudes in a multimode cw dye-laser system have been measured using intracavity absorption spectroscopy. The dependence of the mode-correlation times on the spectral power density in the laser cavity has been investigated in detail. At low spectral power densities, surprising discontinuities are observed and interpreted as dynamical instabilities in the laser system. At high spectral power densities, the mode-correlation time approaches a constant value. These observations are described by a system of two nested, self-referential feedback loops explaining the behavior of the laser system. The resulting model is discussed within the concepts of generalized multistability and presumably chaotic behavior.

## I. INTRODUCTION

It is important for the analysis of nonlinear dynamical systems far from thermal equilibrium (synergetic systems[1]) to treat them by nonlinear methods. A linearization often leads to incomplete results which do not account for the essential qualities of the system under consideration.

For quantum optical systems like lasers which are governed by the semiclassical Maxwell-Bloch equations, Haken[2] has shown the striking analogy with turbulent systems obeying the Lorenz equations.[3] These systems often show instabilities which are formally equivalent with thermodynamical phase transitions if they are continuously driven away from thermal equilibrium. Due to fundamental fluctuation-dissipation relations, stochastic forces are imposed on the dynamical behavior of nonequilibrium systems. These forces give rise to fluctuations which are of purely stochastic character. Furthermore, in certain parameter ranges synergetic systems can behave in a way which is known as deterministic chaos if they have at least three degrees of freedom.[4]

In order to investigate quantum optical synergetic systems theoretically as well as experimentally, two limiting cases can be considered:[5]

1. A system with strong and simplified boundary conditions such as a single-mode laser with an injected external signal or with a saturable intracavity absorber. In this low-dimensional case a numerical analysis should be possible.

2. A system with an infinite number of degrees of freedom (NDF) like a multimode laser with a completely homogeneous gain medium. Systems of this kind could be theoretically treated by asymptotic solutions.

Quantum optical systems of type 1 have often been investigated in recent years. Observations of instabilities and transitions to chaotic behavior have been reported by various groups.[6]

The situation is somewhat different for systems of type 2. Usually dye lasers are considered as laser systems with a predominantly homogeneous gain medium. However, standing-wave linear dye lasers exhibit at least small spatial nonuniformities. Therefore, the condition of a completely homogeneous gain medium can never be *exactly* fulfilled. On the other hand, ring dye lasers as systems without standing-wave effects emit a rather small linewidth compared with linear dye-laser systems. For this reason, in ring dye lasers an upper limit is set for the NDF. Therefore it is obvious that there are fundamental problems in realizing the boundary conditions for systems of type 2 by means of dye-laser systems.

As in the case of single-mode lasers, much work has been reported on the dynamical behavior of multimode laser systems during the last years.[6] In particular, we refer to a paper of Westling *et al.*,[7] who measured the temporal autocorrelation of the total broadband output intensity of a multimode dye laser. At a certain critical laser power, the autocorrelation of the total output intensity changed discontinuously, thus showing the typical feature of a laser instability (nonequilibrium phase transition).

In the present paper we demonstrate a novel method for measuring average correlation times of individual modes instead of the total output intensity of a multimode dye-laser system. This method is based on the technique of intracavity absorption. It uses the concept of finite mode lifetimes in cw laser systems which has been stressed in a recent publication.[8]

In Sec. II a reinterpretation of the finite mode lifetimes as mode-correlation times will be discussed. Furthermore, the self-referential character of multimode laser systems with respect to some of their properties will be elucidated. Section III describes the experimental method for measuring mode lifetimes in a standing-wave multimode dye laser. The experimental results are reported in Sec. IV. They allow a clear distinction between the behavior of the system in different regimes of its spectral power density. The discussion of the results will be carried out within the framework of the theory of synergetic systems.

 

## II. MODE LIFETIMES IN cw MULTIMODE LASERS

### A. Mode lifetimes and mode-correlation times

It is the intention of this section to introduce a new, fundamental interpretation of the finite mode lifetimes in cw laser systems. This interpretation will be especially important in view of the laser as a synergetic system. In this context, the essential parameters of a quantum optical system have been defined by Haken.[1] The pump power is regarded as an external *control parameter* of the system. The mode amplitude or the mode intensity, respectively, serves as an *order parameter* in the sense of Landau.[9] The mode intensity is a measure for the action of the laser system.

With growing pump power the gain medium is driven away from thermal equilibrium, until a critical value is reached beyond which stimulated emission prevails. At this threshold pump power a process occurs which can be described analogous to a second-order thermodynamic phase transition.[10,11]

Above the lasing threshold, the state of the quantum optical system is characterized by the coherence of the emitted radiation, thus representing a high degree of order. In case of a single-mode laser the statistical properties of the emitted light are quite well investigated.[12] In a crude approximation, a single-mode laser system well above the lasing threshold follows Poissonian light statistics:

$$\langle \Delta N \rangle^2 \sim \langle N \rangle \; , \tag{1}$$

where $\langle N \rangle$ is the average photon number (order parameter), and $\langle \Delta N \rangle$ is the average fluctuation of the photon number.

In multimode systems the fluctuations of the emitted light per mode $j$ consist of a superposition of Poissonian and non-Poissonian statistics:[13,14]

$$\langle \Delta N_j \rangle^2 \sim \langle N_j \rangle + \langle N_j \rangle^2 \; . \tag{2}$$

Of course, the latter ones predominate well above the lasing threshold. Furthermore, they are responsible for the limitation of the mode lifetimes in a cw multimode laser system.[8] It should be mentioned that the explanation of non-Poissonian statistics in multimode lasers does not require any pump fluctuations. Non-Poissonian statistics result already from intrinsic quantum-statistical fluctuations (such as spontaneous emission). The fluctuations of the total output power are Poissonian, as in the single-mode case.

From the preceding considerations it is clear that the fluctuating parameters in a multimode laser system are identical with its order parameters (the mean photon numbers or the mode amplitudes). The correlation time of these order parameters is governed by their fluctuations. It is a measure for the coherence properties or, from a different point of view, for the stability of individual modes.

The correlation in one particular mode is interrupted if the mode amplitude is quenched by fluctuations. Since this is exactly the mechanism by which the mode lifetime is defined, it is convincing to identify the mode lifetime with the correlation time of the mode amplitude.

With this interpretation a convenient measurement of mode-correlation times in multimode lasers can be performed. As described in a recent paper,[8] the mean mode lifetime is a quantity which is experimentally accessible by intracavity absorption spectroscopy. The application of this method will be discussed in detail in Sec. III.

### B. Mode lifetimes as self-referential quantities

As Baev *et al.*[15] have derived from the analysis of a rate-equation system, an enhanced mode lifetime $t_{\text{mode}}$ lowers the spectral bandwidth $\Delta\lambda$ of the laser emission according to

$$\Delta\lambda \sim (t_{\text{mode}})^{-1/2} \; . \tag{3}$$

This dependence is due to the nonlinear mode coupling provided by the gain medium. Since those modes which exist in the wings of the gain profile are exposed to a lower net gain than modes which are close to the center of the gain profile, the nonlinear coupling gives rise to a redistribution of the mode intensities. Weak modes deteriorate and strong modes are further amplified. In this sense, one arrives at a time-dependent picture, in which the modes in the wings of the gain profile die out as the mode lifetime advances.

If the total output power $P$ of the dye laser is not changed, the spectral width of the laser emission defines the spectral power density $P/\Delta\lambda$. This implies that the spectral power density increases with increasing mode lifetime $t_{\text{mode}}$.

On the other hand, an increasing spectral power density gives rise to an increasing influence of fluctuations on the intensity of individual modes. As a direct consequence, the average mode lifetime decreases with increasing spectral power density due to the intensity fluctuations in the individual modes.[8]

The mutual relationship between the mode lifetime and the spectral power density can be regarded as a so-called self-referential system. This means that the mode lifetime is finally dependent on itself via the intermediate steps which are schematically illustrated in Fig. 1: An increasing mode lifetime provides a spectral narrowing of the laser emission. Therefore the spectral power density increases and the influence of the intensity fluctuations in individual modes is enhanced. As a result, the mode lifetime will be reduced. This control process prevents the system from spontaneous single-mode operation which would imply an infinite mode lifetime. The self-referential character of the system guarantees that a finite equilibrium value of the mode lifetime is reached at every given power.

Generally, for an increasing spectral power density the mode lifetime has been shown to decrease.[8] However, the mode lifetime as a function of the spectral power density can show additional interesting details, which will be discussed in Sec. IV. These features differ from the overall behavior. Therefore, modifications of the purely stochastic theoretical approach given in Ref. 8 might be extracted from such details.

Of course, in an ideally homogeneous gain medium the number of lasing modes is identical with the NDF of the

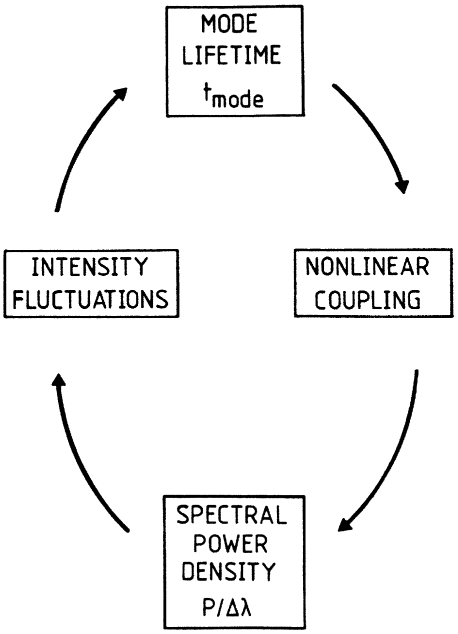


FIG. 1. Schematic diagram of the self-referential feedback loop between the mode lifetime and the spectral power density. The mode lifetime influences the spectral power density via the nonlinear mode coupling. On the other hand, the spectral power density modifies the influence of intensity fluctuations on the lifetimes of individual modes.

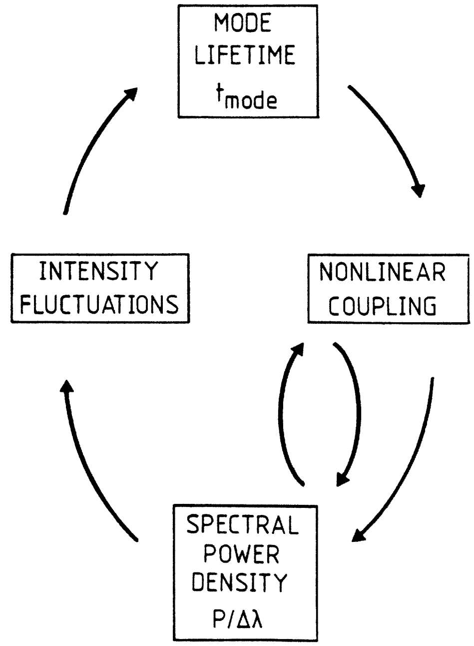


FIG. 2. Extended model of the self-referential system described in Fig. 1. An additional feedback loop between the spectral power density and the nonlinear coupling is introduced. It accounts for discrete changes of the degree of gain inhomogeneities at critical values of the spectral power density.

system. If the gain medium contains inhomogeneous contributions, the coupling between individual modes is enhanced. This agrees with the results of a numerical analysis performed by Brunner *et al.*[16] In case of a strong mode coupling (equivalent with a large inhomogeneous contribution to the gain profile) the NDF of the system can possibly reduce to a small fraction of the number of modes.

The degree of inhomogeneity of the gain medium can be changed by the spectral power density (for example, by spatial nonuniformities). Therefore the control process shown in Fig. 1 has to be extended by an additional feedback loop between the spectral power density and the nonlinear coupling, which is influenced by the inhomogeneity of the gain medium. This nested self-reference is illustrated in Fig. 2. If an increasing spectral power density enhances the inhomogeneity of the gain medium, the coupling between different modes will be changed in the following way.

Let $m$ be the NDF of the system, which is assumed to be identical with the number of *independently* oscillating modes. $M$ is the total number of modes at a fixed spectral power density. Then the ratio $M/m$ is a measure for the inhomogeneity of the gain medium. If the inhomogeneity $(M/m)$ is decreased by an enhanced spectral power density, the number of independently oscillating modes increases. In other words, the nonlinear coupling changes and simultaneously causes a modification of the spectral bandwidth $\Delta\lambda$ and of the spectral power density $P/\Delta\lambda$.

The described process is expected to be initiated only at certain critical values of $P/\Delta\lambda$, because $m$ and $M$ are integers and $M/m$ can only change by a discrete amount. At every critical value of $P/\Delta\lambda$ the system should run through the inner feedback loop shown in Fig. 2. Then a new equilibrium between the spectral power density and the mode lifetime will be established.

The description of the gain inhomogeneity in terms of $M/m$ suggests a picture of $m$ mode packets. They consist of those modes which are strongly coupled, and which oscillate independently from modes belonging to other packets. Thus the system shows cooperative phenomena on two different levels.

(i) The cooperative action of the particles inside the gain medium gives rise to individual longitudinal laser modes.

(ii) Inhomogeneous contributions to the gain medium cause cooperation of individual modes which might result in a reduction of the degrees of freedom from $M$ modes to $m$ mode packets.

## III. EXPERIMENTAL

Mode lifetimes in a multimode cw dye-laser system have been measured by means of intracavity absorption spectroscopy. As recently shown,[8] in the case of an unsaturated absorption the relative depth of an absorption dip $(I/I_0)$ follows a modified Beer's law

$$I/I_0=\exp(-\kappa c t_{\mathrm{mode}} l/L)\ , \quad (4)$$

where $\kappa$ represents the absorption coefficient at the center of the line, $t_{\mathrm{mode}}$ is the average mode lifetime, and $l/L$ is the ratio of the effective absorption length $l$ to the cavity length $L$. $I$ is the laser intensity at the center of the absorption line, $I_0$ is the intensity in the absorption-free case. The mode lifetime $t_{\text{mode}}$ can easily be determined if the absorption coefficient $\kappa$ of the observed line is known.

In order to have convenient experimental conditions, the ubiquitous atmospheric water vapor has been used as a clearly unsaturated absorber. In this case, $l/L=1$. Two different $H_2O$ lines have been investigated which are listed in the following table. The line numbers, their position, and their identification are given according to the work of Antipov *et al.*[17]

| Line no. | Position ($\text{cm}^{-1}$) | Identification |
|---|---|---|
| 198 | 16 852.80 | $P2(401)$ |
| 199 | 16 850.83 | $P2(401)$ |

Both lines belong to the (401) band of the electronic ground state of $H_2O$ and have been selected because they are not blended by other transitions. They are spaced by approximately 2 $\text{cm}^{-1}$. Therefore, they provide independent results for the lifetimes of those two different sets of modes which belong to the corresponding absorption linewidths.

The absorption coefficients of both $H_2O$ lines have been determined as described recently.[8]

A cw jet-stream dye laser with a folded cavity (length $L=60$ cm) has been operated with Rhodamin 6G. The wavelength was tuned by means of an interference filter coated for 450 nm. Apart from the tuning filter, the standard mirrors for Rhodamin 6G were the only optical elements inside the cavity. The absorption features of both molecular transitions were observed under the environmental conditions of the laboratory atmosphere.

The dye laser was operated on a vibration-free table to minimize mechanical instabilities. A laminar flow box covered the whole experimental setup to avoid any perturbing dust particles inside the laser cavity.

The radiation emitted by the dye laser was observed by means of a 2-m grating spectrometer. The spectrum was recorded photoelectrically and registered on a strip-chart recorder using a lock-in amplifier.

An electro-optic modulator between the dye laser and the argon-ion pump laser imposed a fixed mode lifetime between 5 and 15 $\mu$sec on the dye-laser system. Figure 3 shows the measured sensitivity $S=-\ln(I/I_0)$ as a function of the fixed mode lifetime $t_{\text{mode}}$. The absorption coefficients can be directly taken from the slopes of the lines and are indicated in Fig. 3. The absolute water-vapor concentration for this particular measurement has been $(9.81\pm0.34)$ g/cm$^3$ [$N=(3.26\pm0.11)\times10^{17}$ per cm$^3$].

The ratio of the measured absorption coefficients is $\kappa(198)/\kappa(199)=1.97\pm0.12$. This value differs by 12% from the ratio $\kappa(198)/\kappa(199)=1.76$ obtained from the data of Antipov *et al.*,[17] who estimated an error smaller than 10%.[18]

Since the absorption coefficients change due to small variations of the absolute water-vapor concentration, they had to be corrected for each sequence of measurements.

Once the absorption coefficients of the relevant lines

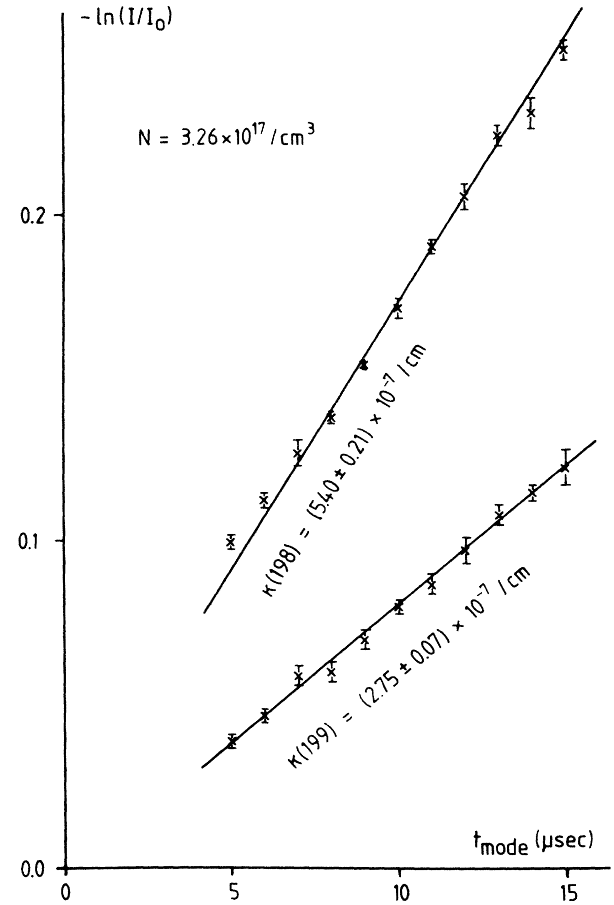


FIG. 3. Measurement of the absorption coefficients in the center of the investigated $H_2O$ lines. Measured sensitivity $S=-\ln(I/I_0)$ of both lines is given as a function of the mode lifetime $t_{\text{mode}}$. The absorption coefficients are calculated from the slope of the lines and the $H_2O$ density $N$.

are known, the mode lifetimes in the free-running system can be extracted from the relative absorption depths $(I/I_0)$ according to Eq. (4). The mode lifetimes have been measured as a function of the spectral density of the dye-laser output power. In order to detect finer details of this dependence, the output power has been incremented in small steps of 10 mW. Each particular mode lifetime has been obtained from five independent measurements of the relative absorption depth of each line.

## IV. RESULTS AND DISCUSSION

The behavior of the mode lifetimes (mode-correlation times) as a function of the spectral power density shows two basically different features. They can be classified according to the spectral power density in the dye laser. At low spectral power densities, we observe discontinuous jumps of $t_{\text{mode}}$ (Sec. IV A). At high spectral power densities, the mode lifetime approaches a constant value (Sec. IV B).

These features modify the picture which results from the purely stochastic approach discussed in Sec. II A. Nevertheless, this approach correctly describes the overall behavior: The mode lifetime decreases with increasing spectral power density.

### A. Discontinuous behavior at low spectral power densities

The mode lifetimes at low spectral power densities are shown in Fig. 4. Their values as extracted from both absorption lines agree within their standard errors. Hence, the given error bars refer to the mean value of both lines.

The spectral power densities as indicated on the abscissa were calculated from the ratio of the dye-laser output power and the spectral width of the laser emission. The intracavity spectral power density can be obtained by accounting for the reflectivity of the output mirror of the resonator.

There are clearly two critical spectral power densities, at which the mode lifetimes show serious deviations from the continuous decrease predicted by theory.[8] At approximately 14.5 mW/Å the mode lifetimes reduce drastically from 300 down to less than 200 μsec. With a further increase of the spectral power density, the mode lifetimes recover to about 240 μsec at spectral power densities around 30 mW/Å. At 36 mW/Å again another discontinuity appears, reducing the mode lifetimes from 240 to 180 μsec.

It has to be pointed out that the critical values of $P/\Delta\lambda$, which give rise to the discontinuities, depend on the particular fine adjustment of the laser resonator. For this reason, the behavior of the mode lifetimes was qualitatively reproduced at different values of $(P/\Delta\lambda)_{crit}$ during subsequent measurements.

In order to illustrate the difference between the behavior of the total output power of the multimode system and its spectral power density as a function of the pump power, we refer to Fig. 5. In the upper diagram, the total output power follows the pump power in a steady and almost linear manner. The slope of the resulting line is a measure for the efficiency of the dye-laser system.

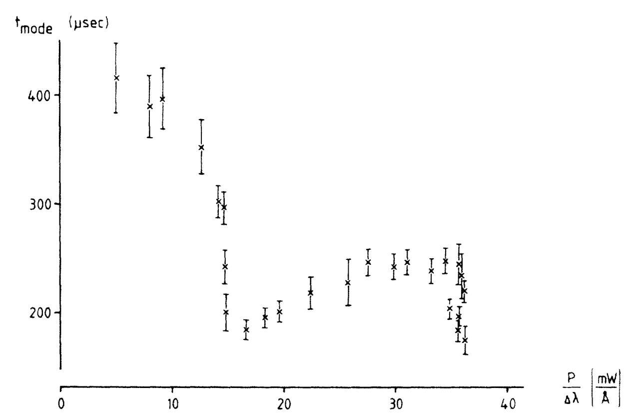


FIG. 4. Measured mode-correlation times as a function of low spectral power densities. The error bars result from a statistical average over both absorption lines. The mode-correlation times show discontinuous jumps at 14.5 mW/Å and at 36 mW/Å in this particular sequence of measurements.

However, the lower diagram shows pronounced steps of the spectral power density in certain pump-power intervals. These steps can be understood phenomenologically by a simultaneous increase of $P$ and $\Delta\lambda$, thus providing an almost constant ratio $P/\Delta\lambda$. The stepwise variation of the slope in the lower diagram of Fig. 5 is directly correlated with the discontinuities of the mode lifetimes in Fig. 4. In those pump power ranges, where the dye-laser bandwidth $\Delta\lambda$ increases with $P$, the pronounced reduction of the mode lifetimes has been observed.

Figure 4 reveals two phenomena which are in contrast to the theoretical predictions based on the stochastic approach mentioned above:

(i) The *discontinuous* reduction of the mode lifetimes at certain critical values of the spectral power density;

(ii) an *increase* of the mode lifetimes, as the spectral power density is further enhanced beyond $(P/\Delta\lambda)_{crit}$.

We start the discussion of these phenomena within the framework of the self-referential properties of the multimode system, as described in Sec. II B.

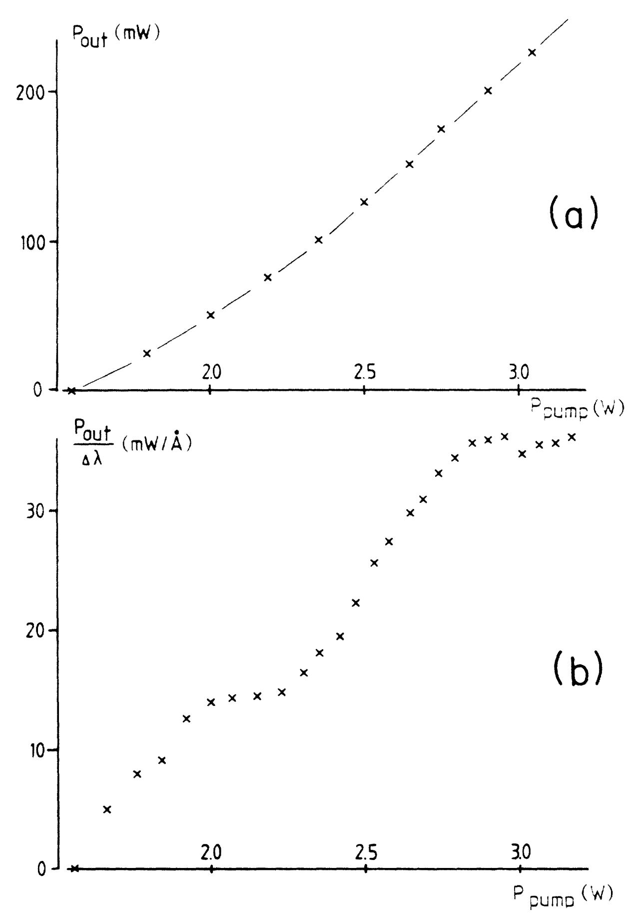


FIG. 5. (a) Output power $P_{out}$ of the dye laser as a function of the pump power $P_{pump}$. An almost linear dependence is obtained, which is equivalent to a constant total efficiency. (b) Spectral power density $P_{out}/\Delta\lambda$ of the dye laser as a function of the pump power $P_{pump}$. In certain pump power ranges, the spectral power density does not change. These particular spectral power densities coincide with the observed discontinuities of the mode-correlation times in Fig. 4.

The nested control processes illustrated in Fig. 2 differ mainly by the fact that the inner feedback loop is expected to be initiated only at certain discrete values of $P/\Delta\lambda$. This assumption is based on the picture of a discrete ratio $M/m$, which is considered as a measure for inhomogeneities inside the gain medium. In contrast to the inner loop, the superior loop connecting the mode lifetime and the spectral power density yields an equilibrium mode lifetime for each laser power $P$.

We argue that the spectral power density reaches a critical value $(P/\Delta\lambda)_{\mathrm{crit}}$ in those regions where discontinuities of the mode lifetime occur. At $(P/\Delta\lambda)_{\mathrm{crit}}$, the degree of inhomogeneity inside the gain medium changes. This variation of $M/m$ causes a modification of the nonlinear coupling among the different modes, thus giving rise to a change of the spectral width of the laser emission. This behavior can clearly be recognized in the lower part of Fig. 5.

With respect to the sudden decrease of the mode lifetime at $(P/\Delta\lambda)_{\mathrm{crit}}$, we are still left with a problem which cannot be solved by stochastic fluctuations as the only mechanism determining the mode-correlation times. Moreover, the stochastic model is also insufficient to explain the growing mode-correlation time with increasing spectral power density beyond $(P/\Delta\lambda)_{\mathrm{crit}}$. Purely stochastic arguments predict a steady decrease of the mode-correlation times.

Consequently, one has to look for concepts which would allow for a modified behavior of the mode-correlation times. In the following, we discuss two different possibilities.

1. Within a potential picture of the laser system, generally some potential valleys may coexist which are separated by intermediate barriers. In a different terminology, it has become customary to speak of coexisting attractors. The system can switch from one attractor to another if the fluctuations of its order parameter are strong enough to drive the system across the separating barrier. This phenomenon has been called "generalized multistability."[5] It can be characterized by a mean first-passage time describing the average time interval during which the system stays in the same attractor.[19] In this sense, the mode-correlation times could be related to mean first-passage times of a multistable system.

2. An alternative concept providing finite mode-correlation times originates from the properties of deterministic chaos. One of these properties is the limited correlation time of an order parameter of the system. As such a quantity, the mode-correlation time could be considered as a measure for the degree of chaos in a multimode laser system. For the system investigated in the present paper, several necessary conditions for chaotic behavior are fulfilled. However, there is not yet enough evidence that the system certainly shows chaotic behavior in distinct parameter ranges.

Without any further experimental information, it is impossible to distinguish between both concepts only on the basis of correlation-time measurements. Nevertheless, there are some indications which suggest an interpretation of the measured mode-correlation times as a superposition of mean first-passage times and chaotic correlation times.

Since the concept of mean first-passage times involves fluctuations (which always exist in real physical systems), it is preferably identified with the overall stochastic behavior of a decreasing mode-correlation time at increasing spectral power density. An increase in the spectral power density is equivalent to an increasing influence of fluctuations due to fluctuation-dissipation relations. This is true as long as the system operates far enough away from dynamical instabilities.

In agreement with results reported by Westling *et al.*,[7] we assume that the multimode laser system is characterized by a considerable gain inhomogeneity at low spectral power densities. This assumption is further supported by the measured mode lifetimes, which are identical (within their statistical errors) for both absorption lines investigated. A rather large gain inhomogeneity is equivalent to only a few independently oscillating mode packets. Hence, the system has only a few degrees of freedom and is of relatively low dimensionality.

Under these conditions, a comparison with numerical results obtained for one-dimensional maps[20] should be justified. The mentioned numerical analysis revealed a linear relation between the logarithms of the first-passage time and the noise amplitude.

In order to compare the numerical results with experimental data obtained for the multimode laser system with only a few degrees of freedom, we fitted a log-log plot of the mode lifetimes versus the spectral power density. This plot is shown in Fig. 6. In the selected range, the behavior of the mode lifetimes qualitatively follows the prediction of the purely stochastic theory. If, to a first approximation, we regard the fluctuation amplitude to be linearly dependent on the spectral power density, we obtain a surprising analogy to the model calculations mentioned above. Our experimental data show an almost perfect linearity according to a correlation coefficient of $-0.991$. They confirm the interpretation of the mode-correlation time by the concept of mean first-passage times, as long as the measured mode-correlation times decrease steadily with increasing spectral power density.

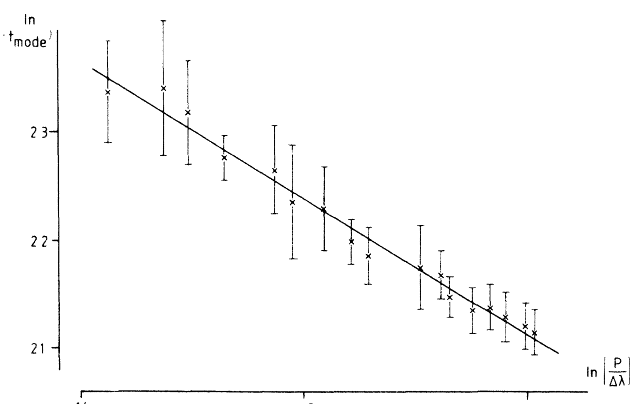


FIG. 6. Log-log plot of the measured mode-correlation times as a function of the spectral power densities $P/\Delta\lambda$. In this case, the selected range of $P/\Delta\lambda$ is free of any discontinuities of the mode-correlation times. The measured values are fitted in a linear manner and reveal a correlation coefficient of $-0.991$.

The discontinuities at critical values of $P/\Delta\lambda$ are interpreted as manifestations of dynamical instabilities of the multimode laser system. The process of symmetry breaking, which is characteristic for such instabilities far from thermal equilibrium, is easily recognized by the discrete change of the gain inhomogeneity $(M/m)$. It simultaneously changes the NDF of the system.

The behavior of the mode-correlation times in the neighborhood of an instability differs seriously from its general behavior which is governed by intrinsic stochastic processes. It will be the subject of further experiments to investigate whether the multimode laser system shows chaotic properties. This would imply an interpretation of the mode-correlation times due to the limited correlation times in chaotic systems. Furthermore, experimental evidence could possibly be extracted for a type of instability which leads from one to another chaotic state of the system. Such kinds of instabilities have already been analyzed numerically for a three-dimensional forced Lorenz system ("chaos-chaos phase transition")[21] and for a one-dimensional map ("crises in chaotic attractors"),[22] respectively. Both investigations reveal a change of the chaotic correlation time in the system if it is driven across the instability region.

### B. Asymptotic behavior at high spectral power densities

At elevated spectral power densities, the mode lifetime decreases and finally approaches a fixed value. This value turned out to be rather accurately reproducible, although the laser system had to be readjusted between different sequences of measurements. For high spectral power densities, we found a limiting mode lifetime $t_{\mathrm{mode}} \simeq (95 \pm 5)$ $\mu$sec. As mentioned above, this $t_{\mathrm{mode}}$ is independent of the fine adjustment of the laser resonator. Hence, it represents *constant* physical properties of the system, which have not changed during the measurements presented in this work. In Fig. 7, the measured mode lifetimes are shown as a function of spectral power densities at elevated values.

The error analysis of the measured mode lifetimes belonging to the two different $H_2O$ absorption lines revealed

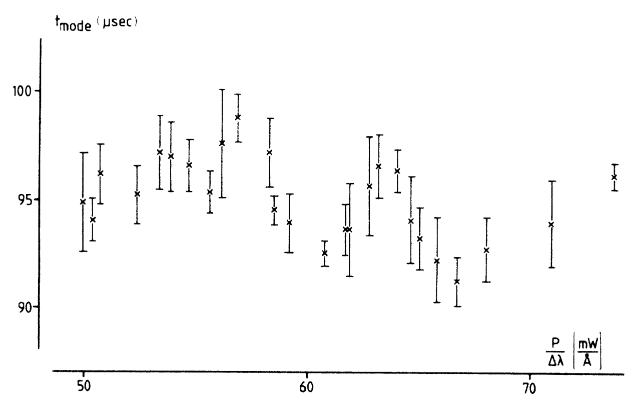


FIG. 7. Measured mode-correlation times at high spectral power densities. As explained in the text, only the values from one absorption line are shown together with their standard error.

the following remarkable fact: Between both particular $H_2O$ lines we obtained differences in the mode lifetimes, which were statistically significant. This result is in contrast to the mode lifetime statistics at low spectral power densities. In the latter case the mode lifetimes agreed within their standard errors.

For these statistical reasons, Fig. 7 shows the mode lifetimes belonging to one of the two $H_2O$ lines only (line 198). Later on we shall focus on the difference between both mode lifetimes resulting from the statistical analysis.

Besides the convergence of the mode lifetimes for high spectral power densities, Fig. 7 shows a feature which can be characterized as an aperiodic oscillation of $t_{\mathrm{mode}}$ as a function of $P/\Delta\lambda$. An identical structure appears if the mode lifetimes around line 199 are considered. This behavior of the mode lifetimes can be interpreted by the action of the self-referential feedback loop between $t_{\mathrm{mode}}$ and $P/\Delta\lambda$.

It would be consistent with the explanation given for the discontinuities at low spectral power densities (Sec. IV A) to identify the local maxima and minima of the mode lifetimes in Fig. 7 with critical values of $P/\Delta\lambda$. Since the influence of fluctuations on the mode amplitudes is large compared with the situation at low spectral power densities, the expected discrete jumps become smeared out.

An additional hint which supports this qualitative picture is given by the small spacings of supposed successive $(P/\Delta\lambda)_{\mathrm{crit}}$. With increasing spectral power density this spacing should decrease if the difference between successive values of $M/m$ decreases. Hence the number $M$ of independently oscillating modes increases with growing spectral power density, thus reducing the degree of gain inhomogeneity. A similar result has been found by Westling *et al.*[7] They argued that the onset of *actual* multimode operation (all modes oscillating independently) occurs beyond a critical power inside the dye laser. In this context we revisit the statistically significant difference between the measured mode lifetimes from different $H_2O$ absorption lines. These different mode lifetimes can be explained by a lowered gain inhomogeneity due to an increased spectral power density. This would imply that the NDF in the standing-wave multimode system increases with an increasing spectral power density.

The fluctuation-dominated character of the multimode laser system at high spectral power densities allows the following interpretation of the mode-correlation time in this regime. In such a situation, fluctuations may be larger than the interior barriers of the laser potential, which has been used to introduce the concept of the mean first-passage time in Sec. IV A. It becomes meaningless to distinguish between different potential wells, since they are bridged by fluctuations: the multistability of the system can be regarded as being degenerate. In the same manner, the concept of an attractor becomes irrelevant. Therefore, the mode-correlation time can certainly not be interpreted as the correlation time of an order parameter in a system, which might possibly be chaotic. As a result, one is left with a purely stochastic interpretation of the mode-correlation times in the case of high spectral power densities.

## V. SUMMARY

In the present paper we introduce a novel method for measuring mode-correlation times in a multimode cw dye laser. This method uses the technique of laser intracavity absorption. The mode lifetime as an important parameter for the dynamical behavior of multimode laser systems is reinterpreted as the correlation time of the mode amplitude.

On the basis of this new concept we measured the mode-correlation time by means of two different $H_2O$ absorption lines in the laser emission spectrum. The observations show a qualitatively different behavior in different ranges of the spectral power density $P/\Delta\lambda$ inside the laser system.

At low spectral power densities, dynamical instabilities occur at critical values of $P/\Delta\lambda$, which depend on the particular fine adjustment of the laser resonator. The decrease of the mode-correlation time with increasing spectral power density due to purely stochastic fluctuations is related to the concept of mean first-passage times in a multistable system. The mode-correlation times around the critical values of $P/\Delta\lambda$ differ strongly from this stochastic behavior. For this situation, the possible relevance of a limited correlation time due to chaotic behavior is still under investigation.

At elevated spectral power densities, the mode-correlation times approach an asymptotic value which is quantitatively reproduced. In this case, fluctuations clearly dominate the behavior of the system. We describe this behavior according to a degenerate multistability. The mode-correlation times are governed by purely stochastic processes.

For the explanation of some of our results, we use a picture of mode cooperation, which is illustrated by mode packets consisting of strongly coupled modes. These modes are not oscillating independently from each other. Their behavior is correlated and can be regarded as an example of self-organization. According to the model described in Sec. IV, the number of independently oscillating mode packets is assumed to increase if the spectral power density is driven across certain critical values. Therefore, the NDF of the system increases with growing spectral power density.

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