Order-disorder transitions in the dynamics of a dye laser

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We analyze the Fresnel number and gain dependence of the spatiotemporal dynamics of a dye laser. The results support the existence of ordered behavior in the low-Fresnel-number and low-gain regions and a transition to spatiotemporal disorder when the Fresnel number and/or the gain are increased. The disordered state has a surprisingly small correlation length (much smaller than the system size), supporting the idea that turbulence is possible in laser systems.

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I. INTRODUCTION

The transition from laminar to turbulent behavior in fluids as one or more control parameters are changed is a well known subject, despite the fact that the concrete mechanisms generating the complex dynamics seen in the turbulent state are not yet completely understood [1]. The fluid example is a classical example, but many other systems have been shown to suffer order-disorder transitions in their spatiotemporal dynamics, leading to complex dynamics and sometimes even turbulentlike behavior [2].

In the field of optics, turbulence has been found in a series of works by Arecchi and others [3], who have investigated defect mediated turbulence and order-disorder transitions in the case of passive systems.

A natural question then arises concerning whether laser devices (the paradigm of coherence and order) are able to show these kind of instabilities. In fact, many laser instabilities have been studied over the past years, but these studies concentrated on plane-wave-type (global oscillations of the spot) [4,5] or on multimode-type oscillations [6–9] where the spatial (transverse) effects do not manifest all their potential diversity. The results of these studies showed good agreement between the experimental data and the simple mathematical models used to describe them, proving that, in those examples, the laser oscillates with a moderate number of transverse degrees of freedom (transverse modes). This is thus an example of the so-called spatiotemporal chaos [10], where there is chaos and some spatial disorder but the transverse correlation lengths are of the order of the system size [11]. However, in a recent work concerning the applicability of the modal expansions it has also been found that multimode expansions with a limited number of terms are not able to explain all the features of the patterns appearing even for not too high Fresnel numbers as stated in [9].

Will the increase of the Fresnel number and/or the gain lead to qualitatively different dynamical states in lasers? Some theoretical studies have addressed this point, but instead of using the complex set of coupled nonlinear equations governing its dynamics (the Maxwell-Bloch equations [12]), they concentrated on better known, simplified equations such as the Ginzburg-Landau equation or the Swift-Hohenberg equation [13], which are expected to retain at least some essential features. The latter work predicted a transition to spatial disorder for moderately high Fresnel numbers. Some fragmentary results are also available for the full Maxwell-Bloch equations, but they are mainly available for the parameter region where multimode behavior is present and in the case of few vortex solutions [14,15]. In this framework accurate numerical schemes [16] with realistic boundary conditions seem necessary to properly address the analysis of more disordered states.

The lack of experimental results, however, has been a limiting factor for the theoretical studies cited before. In a series of studies [17,18] we have analyzed the dynamics of a very high Fresnel number and high gain dye laser and finally proved [19] that its dynamics in the disordered state arising for high Fresnel numbers can be properly called turbulent. This is, as far as we know, one of the first pieces of evidence of a turbulent state in a laser. Despite the disorder found, the dynamics was classified as weak turbulence, not strong. The assumption that filamentary structures are created and interact, leading to "chaos of structures" or weak turbulence, allowed us to derive a simple high-dimensional ordinary differential equation model that allowed a close comparison to the experimental results in the turbulent regimes. However, the existence of these structures was postulated on the basis of previous somewhat related theoretical predictions [20,21] made in very simplified contexts and on the grounds of physical considerations, but no serious demonstration of its existence was given. The similitude of the predictions to the experimental results was also an important support for the validity of the assumptions made, there among the existence of filamented regions.

Other pioneering work in this field has been the experiment of Ref. [22], where the behavior of a high Fresnel number CO_2 laser was studied. However, no theoretical interpretation of the results was made and no characterization of the dynamics was given by the authors. However, they presented very complex patterns and found a disordered state, its dynamics being characterized sometimes by the presence of an unexplained characteristic frequency.

In this work we expand the experimental results of [19]: In the first place, we provide a global picture of the dependence of the dynamics with the Fresnel number and the pumping, which provides (as far as we know, for the first time in laser physics) clear transitions from spatial order to turbulence (not only chaotic multimode oscillations) due to transverse effects. In the second place, a deeper understanding of the local dynamics is achieved by the measurement of the correlation length in the disordered stage, which could be related to the size of the filamented structures if they really exist.

II. EXPERIMENTAL RESULTS

A. Experimental setup

As in the early measurements [17,18] we have worked with a coaxial flash-lamp pumped dye laser with a 15-mm near field cross section diameter and a 500-ns pulse width. With the typical resonator lengths used, the order of the maximum mode excited without internal diaphragms should be around $\mathcal{F} = \frac{b^2}{\lambda L} \simeq 100$, where b is the transverse radius of the resonator, λ is the wavelength of the light, and L is the resonator length. This value greatly exceeds the theoretical prediction of Staliunas [13] for the turbulence "threshold" in the context of a Swift-Hohenberg equation related to the Maxwell-Bloch equations, which is $\mathcal{F} \simeq 20$. The device is then likely to provide turbulent phenomena. For simplicity we will refer to $\mathcal{F}$ as the Fresnel number, although there is a difference of a factor π with other definitions. This number has the more physical meaning of giving an idea of the order of the highest mode that will be excited for the given resonator parameters.

An internal rotatable Brewster window sets the polarization at 45° from the vertical plane. Usually the rear mirror is a plane dielectric total reflector and the output coupler is a 70% transmission plane dielectric mirror, but some measurements were taken with 10% and 40% trasmission mirrors. A polarizing corner cube beam splitter divides the laser beam into two equally intense orthogonally polarized beams (Fig. 1). Each beam runs across a pinhole to fall on the coupling window of an optical fiber cable. Both pinholes are adjustable in diameter

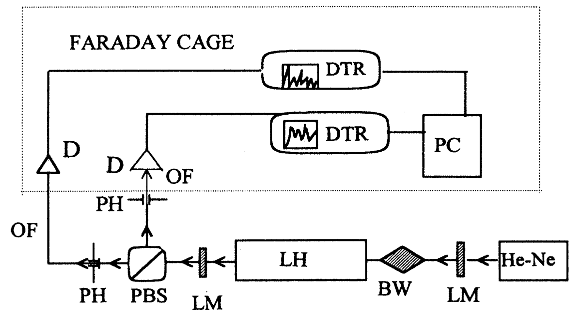

FIG. 1. Experimental setup for measuring correlations. He-Ne, He-Ne alignment laser; LM, laser mirror; BW, Brewster window; LH, laser head; PBS, polarizing beam splitter; PH, pinhole; D, silicon detector; OF, optical fiber; DTR, digitizing transient recorder; PC, personal computer.

and in their spatial location on the laser beam cross section by micrometric screws. Inside a Faraday cage the optical fiber cables are coupled to 1-ns rise time silicon photodiodes, linked to the transient analyzers by a 50-Ω matched coaxial line. We used a two-channel Textronix DSA602 transient digitizer when time resolution was not essential and two fast transient digitizers (a Textronix 7912AD and a Textronix SCD1000) when time resolution was important. The laser strong radio-frequency field noise was shielded by the Faraday cage.

The two pinholes and laser mirrors are aligned by means of an auxiliary He-Ne laser. The system allows the pinholes to be placed within a circle of approximately 0.5 mm diameter around a point in the laser beam cross section. From there, a sweep using a micrometric screw even allowed the placement of the pinholes within a region of about 100 μm around a given point. The system is synchronized to simultaneously register the signals detected in both photodiodes. Afterward, they are subjected to a process as described in [18] to remove the irregular fluctuations from the average behavior. Both fluctuation registers are analyzed on a personal computer. This setup allowed the analysis of correlations of different points of the laser spot.

Using just one of the two regulable size apertures, it

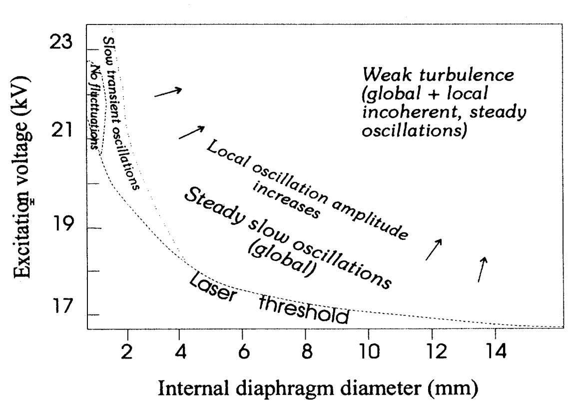

FIG. 2. Global picture of the laser intensity dynamics as a function of the internal diaphragm diameter and the pumping energy (kV).

is possible to measure the dependence of the fluctuation amplitude with the external diaphragm size, which gives a measure of how local the dynamics is.

B. Dependence of the dynamics on the Fresnel number and gain variations

In this experiment we qualitatively analyzed the dynamics to give an insight on the different dynamical regimes appearing for different values of the pumping and Fresnel number. The Fresnel number was changed by placing internal diaphragms from 15 mm (the size of the laser tube), which leads to a Fresnel number $\mathcal{F} \simeq 100$, to 1.5 mm, which corresponds to $\mathcal{F} \simeq 1$. The fact that the dye laser resonant wavelength is very small is decisive for obtaining such very large Fresnel numbers, which are almost impossible to obtain with the CO_2 laser and other

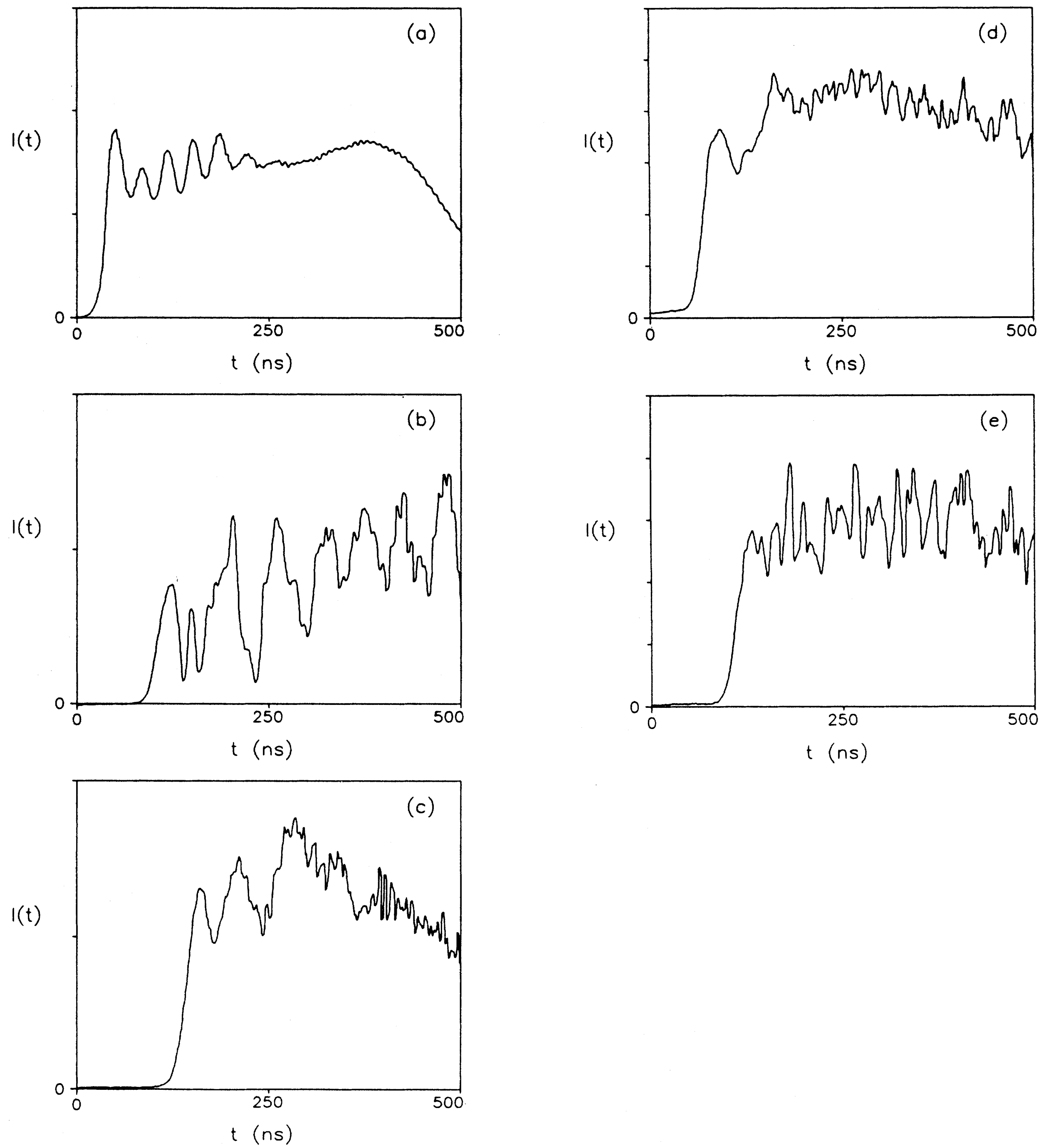

FIG. 3. Typical laser pulses for different control parameter values: (a) $V = 23$ kV, $\phi = 1.5$ mm; (b) $V = 23$ kV, $\phi = 3$ mm; (c) $V = 23$ kV, $\phi = 4$ mm; (d) $V = 23$ kV, $\phi = 10$ mm; (e) $V = 23$ kV, $\phi = 15$ mm.

far infrared lasers commonly used in nonlinear dynamics experiments. Due to the pulsed nature of the laser, we could achieve a very large amplification, much larger than the one obtained with continuous lasers.

Although a global picture can be found, very exact values for the transitions or a very detailed analysis of a particular route cannot be done with our setup due to various reasons: First, the degradation of the dye and that of the flashlamp must be taken into account. Although the dye was changed frequently, an effect in the gain due to the degradation it is unavoidable. Second, the alignment conditions influenced the results and finally the temperature of the dye was also a noise factor influencing the observed dynamics. Despite these limiting factors, we have been able to obtain global qualitative information on the different dynamics present in the laser under study.

We summarize the global picture in Fig. 2. The region on the left is the usual nonlasing region where only fluorescence is emitted because the pumping does not reach the first threshold. The different dynamical behaviors found experimentally are shown in the figure. To clarify the meaning of the different regimes we will analyze in detail one particular case corresponding to a fixed (high) pumping value V = 23 kV and different internal diaphragm diameters ranging form ϕ =1.5 to 15 mm.

As can be seen, in the low-Fresnel-number regime the local dynamics are very simple leading to a transient slow oscillation [Fig. 3(a)]. The slow oscillations found in this regime have simple averaged Fourier spectra [Fig. 4(a)] with a slow dominating frequency of around 16 MHz [a small contribution of the second harmonic is also evident in the spectrum shown in Fig. 4(a)] and are transversely correlated along the laser spot as is evident in (Fig. 5), where the time signals on two different points of the laser spot are presented for a similar case (with a somewhat lower pumping) being clear that the oscillation is synchronous between different points.

When the internal diaphragm is increased to ϕ = 2 mm, the Fresnel number increases. The dynamics becomes somewhat more complex: in the first place, the oscillations become stationary [see, for example, Fig. 3(b)];

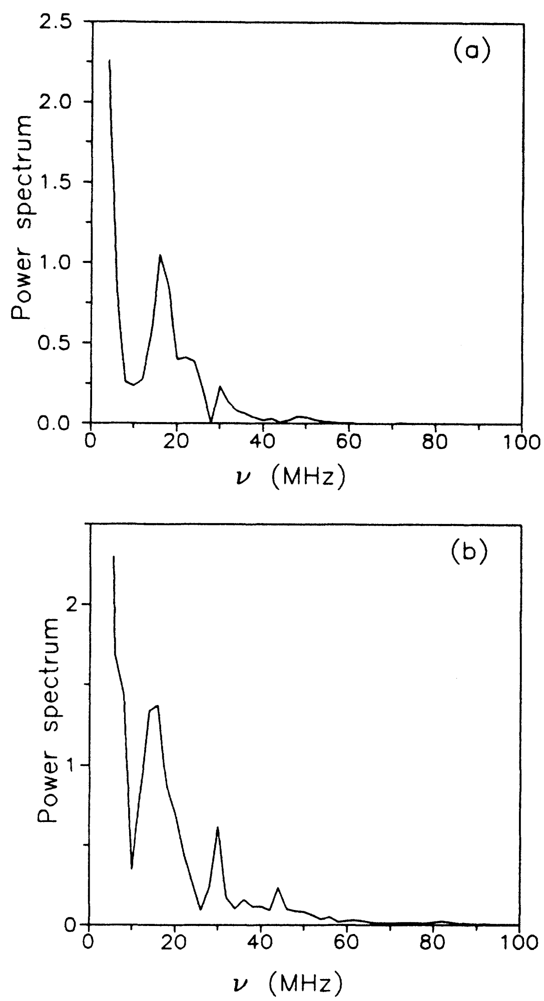

FIG. 4. Averaged power spectrum for (a) ϕ =1.5 mm, V = 23 kV and (b) ϕ =2 mm, V = 23 kV.

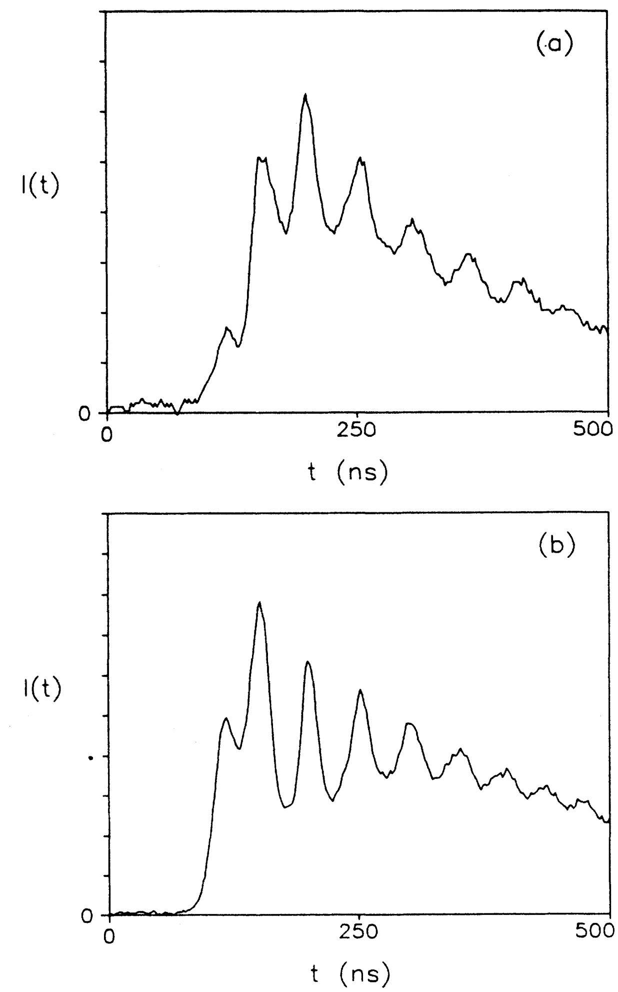

FIG. 5. (a) and (b) Simultaneous local dynamics on two different points on the laser spot separated by approximately 2 mm for pumping level V= 22 kV and internal diaphragm ϕ = 2 mm.

second, the harmonics of the fundamental frequency play a more important role [see the differences in the dominating harmonics between Figs. 4(a) and 4(b)].

A further increase of the internal diaphragm size to $\phi = 4$ mm leads to the appearance of a faster disordered oscillation [Fig 3(c)]. These fast disordered oscillations become more and more dominant as the Fresnel number is increased, as is clear from Figs. 3(d) and 3(e), where single shots for $\phi = 10$ mm and $\phi = 15$ mm are presented.

The fact that the fast oscillation are uncorrelated is clear when we detect the laser intensity on two different points of the laser spot. In Fig. 6 the fluctuations on points separated by 2 mm are presented for $V = 22$ kV and $\phi = 15$ mm. This decorrelation was analyzed in [19] and found to be caused by weak turbulence.

It is interesting to point out that the slow oscillation survives when the local fluctuations appear. In Fig. 7(a) a local register for $V = 23$ kV, $\phi = 15$ mm is presented and in Fig. 7(b) the fast component and the slow part are separated using the filter described in [18]. The fast oscillation has a slow envelope, the envelope frequency being that of the slow oscillations found for low Fresnel

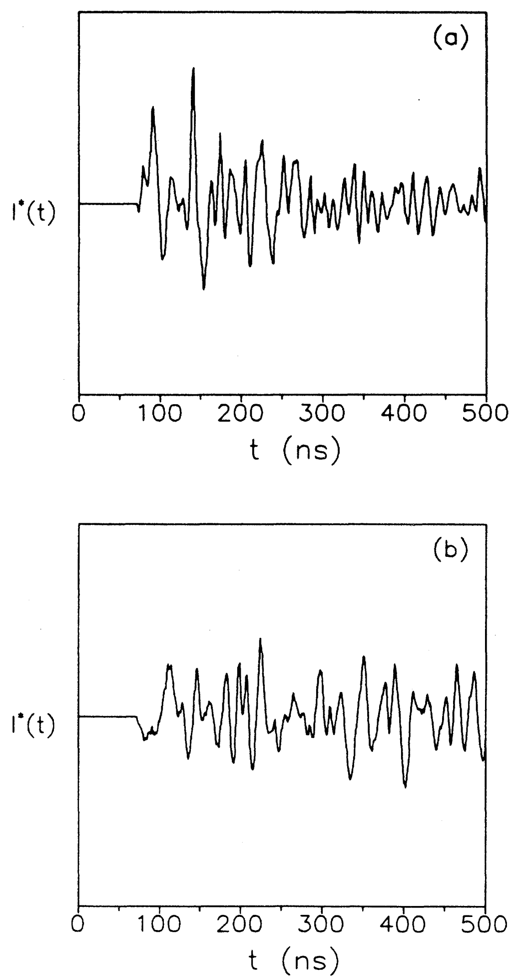

FIG. 6. Synchronously measured local intensity fluctuations on two different points of the laser spot for V =22 kV, ϕ = 15 mm.

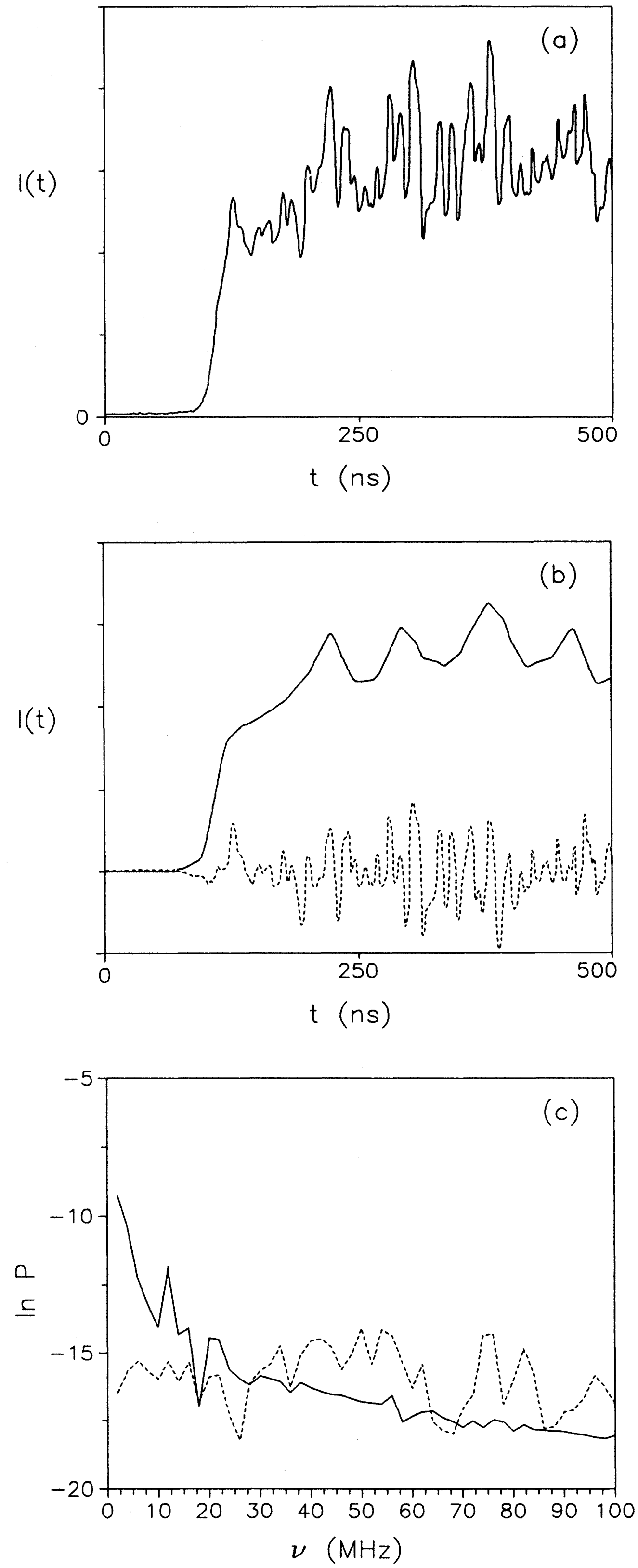

FIG. 7. (a) Local intensity fluctuations for V=23 kV, ϕ=15 mm. (b) Separation of the fast and slow components of the pulse. (c) Power spectra of fast and slow parts.

numbers. In this regime, however, the slow oscillation is also spatially uncorrelated because as the area of the spot registered in the photodetector is increased, both the fast and slow oscillations average and give a smooth nonoscillating global intensity profile. However, the decorrelation of the slow oscillation is not as drastic as that of the high frequency component, as it will be shown later. Figure 7(c) shows the Fourier spectra for both the fast and the slow components of this pulse. The slow evolution frequency component is very simple, leading to a clear peak that corresponds to the inverse of the observed period while the fast frequency spectrum is broadband centered around 50 MHz, which is the characteristic averaged frequency of the fast oscillations.

The fact that the slow component survives is also clear from Fig. 8, where the power spectrum for decreasing internal diaphragms is shown. The disorder-order transition is clear, supporting the picture presented before. A remarkable point concerning the slow oscillation is that *it is not a global relaxation oscillation.* This affirmation is supported by the fact that the relaxation frequency is very dependent on the losses, pumping level, and detuning. We have changed many physical parameters directly related to these quantities and the basic slow oscillation frequency did not change. The voltages selected were varied from threshold to 23 kV and different reflectivities were used for the output mirror: namely, 30%, 60%, and 90%, without appreciable change on the base slow frequency value. Neither the introduction of internal diaphragms nor the change of the resonator length seems to affect significantly the *frequency* of these oscillations. The conclusion that they are of a different dynamical nature than the relaxation oscillations is unavoidable. Also they appear mainly in the low-Fresnel-number regime, which points to a relationship to some kind of mode competition or a related phenomenon. But it is not a mode beating frequency. In our resonator the longitudinal mode beating frequency must be of the order of 150 MHz, which is much faster than the observed oscillation. On the one hand, in a plane to plane resonator the transverse modes are expected to be degenerated and a transverse mode beating cannot appear, at least with such fixed frequency. On the other hand, the fast oscillation has a clear dynamical origin and seems to be generated by the coupling of laser condensates (polarization filaments), as discussed in Ref. [19]. The slow oscillation cannot be explained by this mechanism because it is already present in the ordered region, where filamented structures should not exist.

If we consider the effect of internal losses in the dye a new term arises in the Maxwell-Bloch equations, as discussed in [5], which can introduce a new characteristic frequency in the dynamics. We have checked that this frequency does not generate intensity pulsations in the parameter region under consideration so that this is not the origin of the observed oscillations.

In conclusion, we are not able to find a clear origin for this low-Fresnel-number, slow oscillation. We think it may be of a dynamical nature and should arise from a detailed analysis of the Maxwell-Bloch equations. May be it is connected with the field diffusion induced by the

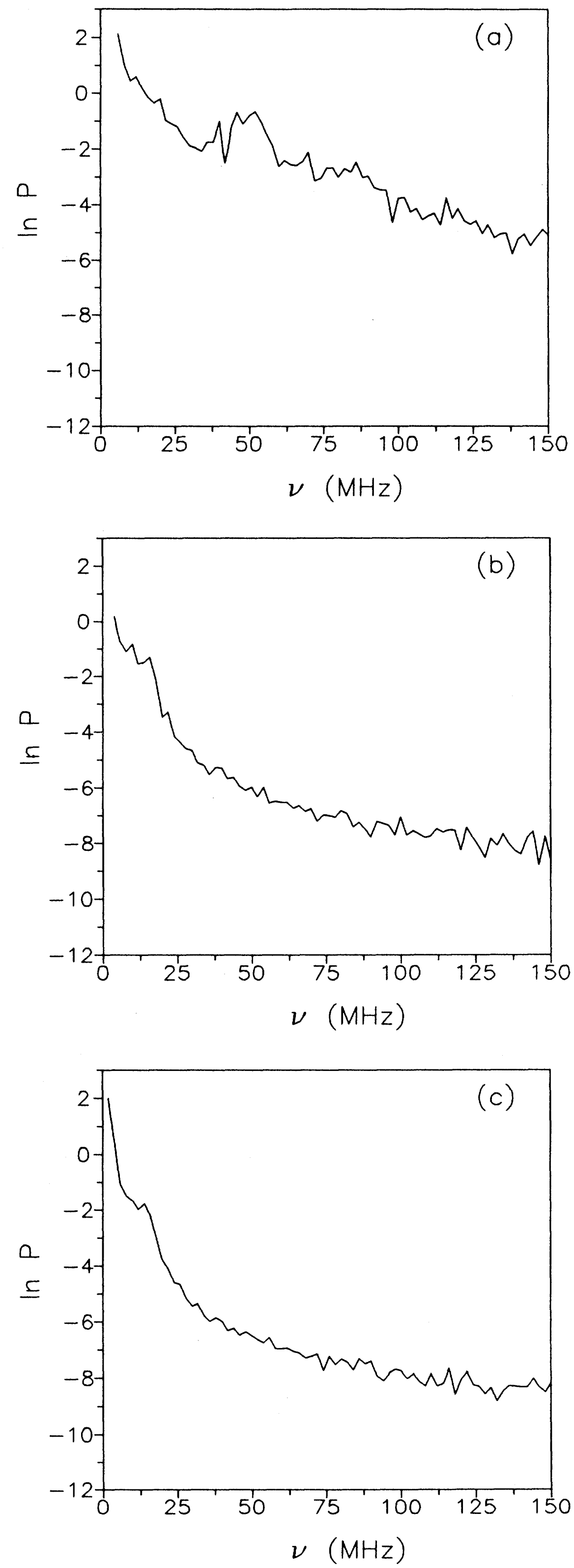

FIG. 8. Change on the averaged logarithmic power spectrum under internal diaphragm diameter variations for $V = 20$ kV. (a) $\phi = 15$mm, (b) $\phi = 6$ mm, and (c) $\phi = 4$ mm.

internal losses of the dye or any other mechanism related to the transverse structure [23]. Perhaps the strange low frequency appearing in the dynamics of the CO_2 laser studied in Ref. [22] is of a similar nature.

C. Correlation lengths in the turbulent state

In the framework of simple energy considerations Emelyanov and Yukalov [20] predicted that a laser medium should tend to develop a filamentary structure. In their elegant paper they obtained an estimation for the size b of those filaments, which is

$$b = 0.22\sqrt{\lambda L}, \tag{1}$$

with λ the laser wavelength and L the resonator length. If we compute it for our typical resonators lengths, we obtain an estimate of $b \simeq 0.22\sqrt{\lambda L} \sim 100\ \mu$m.

In our previous work [19] we were not able to measure the correlation length in the disordered state, but used the information of the decay of the amplitude of the fluctuation (and other evidence) to infer that the disorder was not complete and a finite transverse correlation length related to the size of the characteristic structures exists. We assumed the existence of a correlation length in this range and then used the assumption that the structures existed to develop a theory of diffraction coupled filaments, which explained the dynamical aspects of the turbulent state.

Do those filamentary coherence structures really exist? The answer to this question is related to the other ones: What is the correlation length? Is it in agreement with the predictions of filament-formation theories?

We have been able to perform this measurement using smaller areas than before by using very small calibrated pinholes down to a 10 μm diameter and found [Fig. 9(a)] that the correlation length in the fully disordered state is of the order of 30 μm, which is very small. This size should be connected to the extension of the coherence filaments discussed in [19]. An enhancement

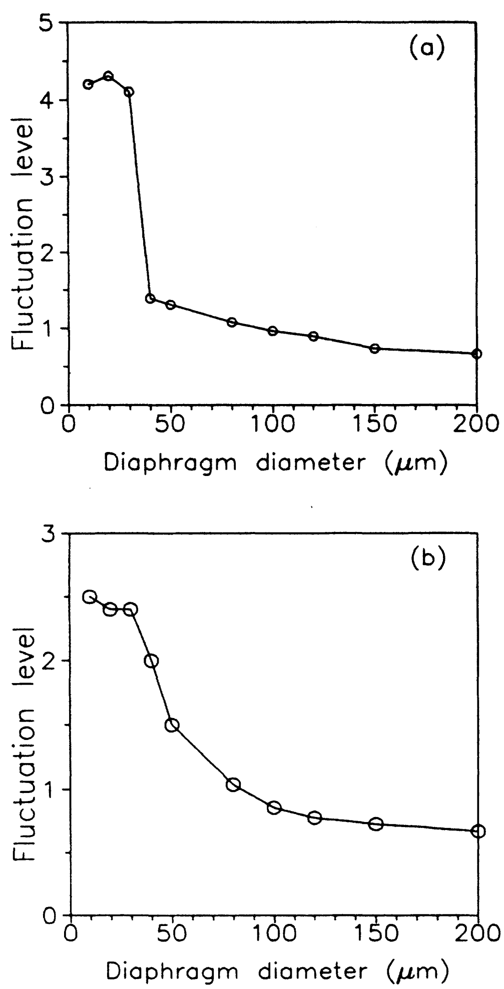

FIG. 9. Experimental dependence of the relative amplitude of the irregular fluctuations on the external diaphragm diameter (which determines the size of the registered region of the laser spot). (a) $V = 20$ kV, $\phi = 15$ mm and (b) $V = 22$ kV, $\phi = 4$ mm.

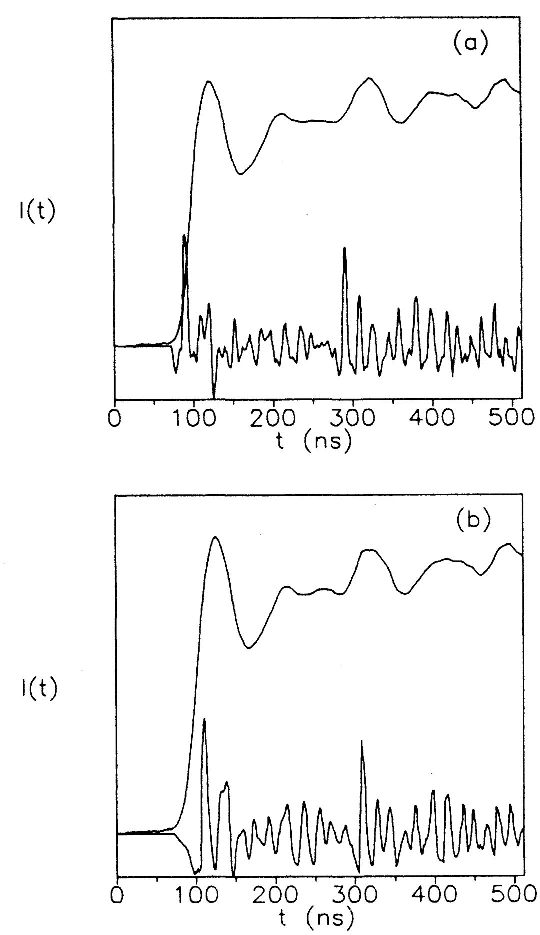

FIG. 10. Fast and slow components of two local intensities belonging to the same pulse located on two regions separated around 100 μm. The slow evolution is correlated while the fast is uncorrelated. The pulse is taken with $V = 20$ kV, ϕ=15 mm.

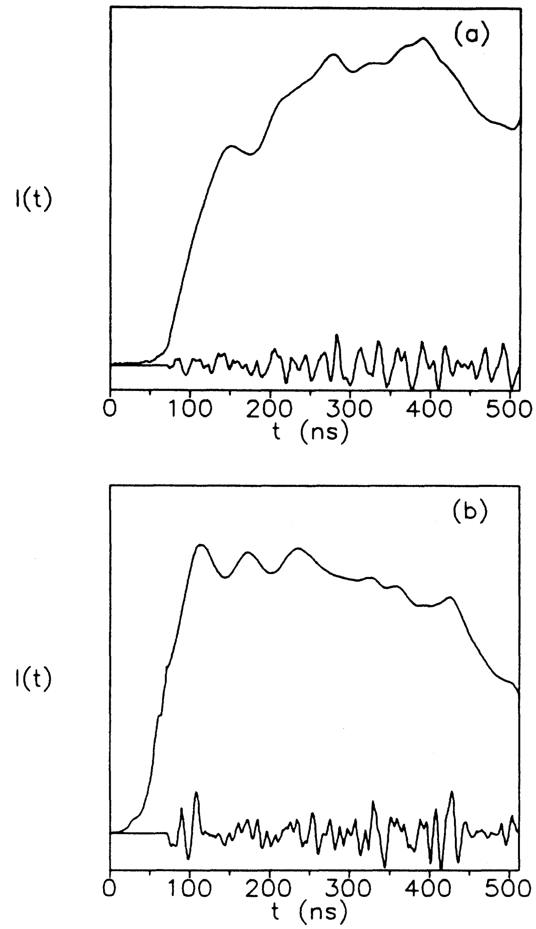

FIG. 11. Fast and slow components of two local intensities belonging to the same pulse located on two regions separated around 200 μm. Both the slow and fast evolution are uncorrelated. The pulse is taken with $V = 20$ kV, ϕ=15 mm.

in the transverse correlation length is observed when the Fresnel number is lower, but still corresponding to spatiotemporal disorder [Fig. 9(b)]. Let us note that not only is the width of the peak somewhat larger (being around 40 μm in the second case), but also the asymptotic decay of the amplitude fluctuation is less abrupt in the lower-Fresnel-number case. If the structures conjectured in Ref. [19] really do exist, this means that the average number of structures on a 15-mm-diam spot could be around 5×10^5. For a 2-mm-wide spot the number of structures is probably not too high because of the tendency of the correlation length to grow when the Fresnel number is decreased. Let us note that the simple estimate of Ref. [20] is reasonably good, despite the crudeness of the approximations necessary to obtain formula (1). We have also checked that the fluctuation amplitude grows when the resonator length is increased, which may be related to the larger size of the domains predicted by Eq. (1) for that case, which should lead to less averaging.

Concerning the low frequency dynamics, it is clear from our previous statements that it is globally correlated in the low-Fresnel-number regime and uncorrelated in the high-Fresnel-number regime; however, the decorrelation is not high as it happens with the fast Fourier components. Figure 10 shows a register for two points separated by 100 μm, where it is clear that the slow oscillation are very similar. When we go 100 μm further the dynamics are clearly uncorrelated (Fig. 11). This behavior is also clear when averaging because upon external diaphragm increases, it is observed that first the fast frequency component disappears and finally the slow component averages and gives smooth nonoscillating pulse shapes when large areas are detected (when more than $\phi = 200$ μm of external diaphragms are used).

III. CONCLUSIONS

The transitions order chaos in laser dye dynamics under some parameter variations have been studied in some detail. As it was expected, the Fresnel number (which measures the separation of transverse degrees of freedom) and the gain (which measures the number of excited degrees of freedom) play a key role in the order-turbulence transitions as predicted in recent theoretical works. The existence of a new oscillation present in the low-Fresnel-number regime has been reported, although its physical origin is not clear yet.

The measurement of the correlation length by means of an indirect procedure has allowed a comparison with the predictions for laser coherence filamentation, which has been the starting point for other works in the area and has been found to be qualitatively correct. The order-disorder transitions reflect also on the behavior of the correlation length when the Fresnel number is decreased.

The classical image of lasers as a paradigm of order and coherence has to be considered with care because of the existence of such disordered states that have more to do with the complex behavior of a turbulent fluid than with the peaceful ordered behavior of laminar ones.

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[1] *Hydrodynamic Instabilities and the Transition to Turbulence*, edited by H. W. Swinney and J. P. Gollub, Topics in Applied Physics Vol. 45 (Springer-Verlag, Berlin, 1985).
[2] M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 3 (1993).
[3] F. T. Arecchi, G. Giacomelli, P. L. Ramaza, and S. Residori, Phys. Rev. Lett. **65**, 2531 (1990); *ibid.* **67**, 3749; F. T. Arecchi, S. Bocaletti, P. L. Ramaza, and S. Residori, *ibid.* **70**, 2277 (1993).
[4] N. B. Abraham, L. A. Lugiato, and L. M. Narducci, J. Opt. Soc. Am. B **2**, 7 (1985), and references cited therein.

[5] I. Pastor, V. M. Pérez-García, F. Encinas-Sanz, J. M. Guerra, and L. Vázquez, Physica D **66**, 421 (1993).
[6] L. A. Lugiato, G. L. Oppo, L. M. Narducci, J. R. Tredicce, and M. A. Pernigo, J. Opt. Soc. Am. B **7**, 1019 (1990).
[7] M. Brambilla, F. Battipede, L. A. Lugiato, V. Penna, F. Prati, C. Tamm, and C. O. Weiss, Phys. Rev. A **43**, 5090 (1991).
[8] J. Tredicce, E. J. Queel, A. M. Gazzawi, C. Green, M. A. Pernigo, L. M. Narducci, and L. A. Lugiato, Phys. Rev. Lett. **62**, 1274 (1989).
[9] M. Brambilla, M. Cataneo, L. A. Lugiato, R. Pirovano, F. Prati, A. J. Kent, G. L. Oppo, A. B. Coates, C. O. Weiss, C. Green, E. J. D'Angelo, and J. R. Tredicce, Phys. Rev. A **49**, 1427 (1994); A. B. Coates, C. O. Weiss, C. Green, E. J. D'Angelo, J. R. Tredicce, M. Brambilla, M. Cataneo, L. A. Lugiato, R. Pirovano, F. Prati, A. J. Kent, and G. L. Oppo, Phys. Rev. A **49**, 1452 (1994).
[10] M. Caponeri and S. Ciliberto, Physica D **58**, 365 (1992).
[11] L. M. Narducci, G. L. Oppo, J. R. Tredicce, L. A. Lugiato, F. Prati, in *Coherence and Quantum Optics VI*, edited by J. H. Eberly *et al.* (Plenum, New York, 1990).
[12] H. Haken, *Laser Theory* (Springer-Verlag, Berlin, 1970).
[13] K. Staliunas, Phys. Rev. A **48**, 1573 (1993).
[14] P. Coullet, L. Gil, and F. Rocca, Opt. Commun. **73**, 403 (1989).
[15] E. D'Angelo, C. Green, J. R. Tredicce, N. B. Abraham, S. Balle, Z. Chen, and G. L. Oppo, Physica D **61**, 6 (1992).
[16] *Title*, Vol. 329 of *NATO Advanced Study Institute, Series B: Physics,* edited by K. H. Spatschek and F. G. Mertens (Plenum, New York, 1994), pp. 287–298; I. Martín, Víctor M. Pérez-García, J. M. Guerra, F. Tirado, and L. Vázquez, in *Fluctuation Phenomena: Disorder and Nonlinearity*, edited by A. R. Bishop, S. Jiménez, and L. Vázquez (World Scientific, Singapore, 1995), pp. 390–396; R. Indik and A. C. Newell, Math. Comput. Simul. **37**, 385 (1994).
[17] I. Iparraguirre and J. M. Guerra, Opt. Commun. **67**, 293 (1988).
[18] I. Pastor and J. M. Guerra, Appl. Phys. B **51**, 342 (1990).
[19] V. M. Pérez-García and J. M. Guerra, Phys. Rev. A **50**, 1646 (1994).
[20] V. I. Emelyanov and V. I. Yukalov, Opt. Spectrosc. **60**, 634 (1986); A. V. Andreev, V. I. Emelyanov, and Yu. A. Iinskii, *Cooperative Effects in Optics* (IOP, Bristol, 1992).
[21] S. Diachenko, A. C. Newell, A. Pushkarev, and V. E. Zakharov, Physica D **57**, 96 (1992).
[22] D. Dangoisse, D. Hennequin, C. Lepers, E. Louvergnaux, and P. Glorieux, Phys. Rev. A **46**, 5955 (1992).
[23] O. Gómez-Calderón, V. M. Pérez-García, J. Lega, and J. M. Guerra (unpublished).