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INSTABILITIES AND CHAOTIC BEHAVIOR OF ACTIVE AND
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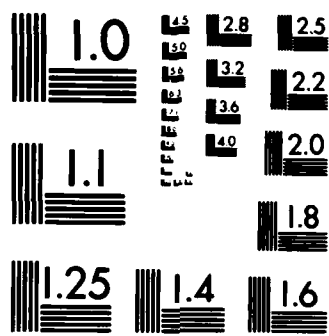
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The main thrust has been to investigate the occurrence of unstable behaviors in active and passive quantum optical systems. Considerable theoretical progress has been made with optical bistability to the point that no additional efforts appear to be

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needed on this front. Research is still very active, instead, to understand the origin of pulsations and chaos in lasers with an injected signal, laser systems with a nonuniform transverse intensity profile, and inhomogeneously broadened high-gain lasers.

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INSTABILITIES AND CHAOTIC BEHAVIOR OF ACTIVE AND PASSIVE LASER SYSTEMS

A final report prepared for the
U.S. Army Research Office

by

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INTRODUCTION

A common trait of matter in thermal equilibrium is uniformity on the large scale and fluctuations on the level of matter's elementary building blocks. This state of perfect democracy, where no single microscopic or macroscopic mode of motion is preferred, is controlled by the second law of thermodynamics and maintained by dissipation and irreversibility. If our world were at thermal equilibrium on a local scale, no structures could survive or develop a hierarchy of purposeful components.

Spatial and temporal structures, on the other hand, do exist in the world around us in many forms, from the complex cell differentiation of living organisms to the highly regular heartbeat of man-made atomic clocks, to the coherent oscillations of a well-stabilized laser.

Over the last several years, a large effort has been underway to understand the conditions that favor organized behavior at a macroscopic level. Quantum optics, the science that gave us the laser and all its applications, has been an active contributor in the search for the general principles that govern structured macroscopic phenomena. The theory and the observation of self-pulsing in high gain lasers, optically bistable systems and other driven optical devices have suggested general rules to explain the occurrence of organized evolution which are nicely supported by rigorous mathematical considerations. At present, there is strong evidence that structures evolve when a sufficiently nonlinear system is driven away from thermal equilibrium. These structures often develop into progressively more complicated temporal and spatial patterns, each with well-defined domains of stability in control

parameter space; sometimes chaos emerges, even without the assistance of external noise sources.

The appearance of chaos in deterministic models of dynamical systems was totally unexpected in the early 1960's when it was first observed in numerical studies of weather predictions. After all, conventional wisdom would appear to require that solutions of differential equations corresponding to well-defined initial conditions should display certain well-defined regularities and reproducibility features. This intuitive notion is inaccurate, in general, as recent research in the mathematical properties of nonlinear differential equations has shown. In simple terms, the situation can be summarized as follows. The long-term behavior of a dynamical system is controlled by the attractors of the differential equations. Some of these attractors, already well-known from elementary calculus, are the steady states of the system, i.e., configurations such that all the time derivatives of the dynamical variables vanish. These steady states, foci, nodes, saddle points, etc., can be stable or unstable; they can also disappear or reappear as the control parameters of the system are varied. Thus, a stable node can become a stable focus, or a stable focus can become unstable etc., as one "turns the knobs" that govern the evolution.

In addition to these steady states, dynamical systems can also be characterized by periodic solutions whose evolution in the phase-space of the variables corresponds to closed loops (limit cycles). Solutions of this type display characteristic fundamental frequencies (the reciprocal of the round-trip time around the limit cycle) and usually a host of harmonic components, whose origin is easily traced back to the nonlinearities of the system.

By varying the control parameters, the limit cycles change their shape, usually in a continuous way; sometimes, however, qualitatively different solutions emerge. A simple loop may suffer a sort of fission process and split into a longer, simply connected loop, whose fundamental period is twice as long as the original one (period-doubling bifurcation); alternatively, a limit cycle may transform itself into a toroidal surface in two dimensions. Incommensurate pairs of frequencies are likely to appear in this case, corresponding to the two principal directions of travel of the phase-space trajectory on the surface of the torus, and this results into quasi-periodic temporal pulsations with a superficial resemblance to Lissajous figures with incommensurate frequencies.

If a two-dimensional torus becomes unstable, a third incommensurate frequency may appear. Under certain conditions, the surface of the new attractor that has replaced the old toroidal surface acquires a very complicated topology as if an enormous amount of valleys and ridges were shaped on it. Attractors of this type possess very strange properties and for this reason have been labelled "strange attractors". One of the most remarkable features of a strange attractor is the extreme sensitivity of a solution to noise (even numerical noise). Thus, not only will the temporal oscillations usually look random, as the phase space path unwinds on the "rough" surface of the attractor, but even minute changes in the initial conditions will cause major alterations of the calculated solutions.

This rich variety of scenarios is remarkably well matched by the behavior of real life systems in a growing number of situations, not only in physics, but also in chemistry, biology, and many branches of engineering sciences. Attempts have been made to explain certain aspects of sociological, economical

and ecological phenomena in terms of mathematical models of this type. To what extent these observed behaviors are the result of general laws, universal principles--if we want to be more ambitious--is not known at present. The field of laser physics, in a sense, provides an almost unique setting for the analysis of these deep questions: the theory takes its premises from fully microscopic equations and in a clear mathematical fashion, allows the derivation of equations of motion for all the relevant macroscopic variables. Stable and unstable steady states, periodic, aperiodic and chaotic solutions can be analyzed in detail and their origin traced to experimentally accessible control parameters. Ultimately, most theoretical predictions can be tested under laboratory conditions which do not require extreme efforts to match even some of the most idealized theoretical settings.

The main thrust of our efforts over the last several years has been to investigate the occurrence of unstable behaviors in active and passive quantum optical systems. Considerable theoretical progress has been made with optical bistability to the point that no additional efforts appear to be needed on this front. Research is still very active, instead, to understand the origin of pulsations and chaos in lasers with an injected signal, laser systems with a nonuniform transverse intensity profile, and inhomogeneously broadened high-gain lasers. In addition to these areas of research, we have also been involved with the theoretical modelling of some recent laser instability experiments carried out by C.R. Stroud and collaborators at the Institute of Optics in Rochester, New York, with the study of coupled logistic maps and chaos in discrete dynamical systems, with some aspects of the free electron laser problem with multiphoton vibrational excitation of molecules, and, finally, with the experimental realization of a two-photon

laser amplifier.

The main body of this report will be devoted to a brief summary of the status of each project and to an overview of other major activities by the Principal Investigator. A listing of publications for which research was partly supported by the U.S. Army Research Office, and of the researchers and staff involved in this work is also included.

1. OPTICAL BISTABILITY

a) Bistability in a hybrid system with delay.

A bistable optical system driven by a constant input field can develop instabilities and display output oscillations if the feedback mechanism of the device has a built-in delay mechanism. The oscillations can be regular or chaotic as originally pointed out by Ikeda¹ in the first contribution dealing with optical chaos. Experimental evidence of the effect was first provided with a hybrid electro-optic device in which the delay of the feedback loop was made considerably larger than the response time of the system². Additional experimental evidence of self-pulsing and chaotic behavior was also provided by Okada and Takizawa³ with considerably shorter delay times.

Our investigations were prompted by the appearance of significant differences in the observed pulsations of Refs. 2,3 and the results of a theoretical analysis of the problem by Gao, et al.⁴ were already discussed in a previous progress report.

In our follow-up work, we have developed new methods of analysis for the study of the output oscillations based on the power spectra of the output intensity. We have shown that the spectral analysis coupled to a precise knowledge of the complex eigenvalues of the linearized problem and

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1. K. Ikeda, Opt. Comm. 30, 257 (1979); K. Ikeda, H. Daido and O. Akimoto, Phys. Rev. Lett. 45, 709 (1980).
 2. H.M. Gibbs, F.A. Hopf, D.L. Kaplan and R.L. Shoemaker, Phys. Rev. Lett. 46, 474 (1981).
 3. M. Okada and K. Takizawa, J. Quant. Electron. QE17, 2135 (1981).
 4. J.Y. Gao, J.M. Yuan, L.M. Narducci, Opt. Comm. 44, 201 (1983).

of the time-dependent solutions provides a very sensitive tool for describing most features of interest of this system. In our study, we have investigated a wide range of delay values and gained a qualitative understanding of the origin of the periodic and chaotic oscillations. We have also clarified the relation between the short and long delay limits of the bistable model.

Our findings include strong evidence in favor of the fact that the route to chaos involves an infinite sequence of period doubling bifurcations for most values of the delay time, a proof that external noise (e.g., of instrumental origin) is a plausible cause of the observed bifurcation gaps, and an intriguing theory that three or more independent frequencies may coexist without the system developing a strange attractor. *Additional technical details can be found in Papers 1 and 2 listed at the end of this report.*

b) Bistability in a total internal reflection optical cavity

A few years ago Kaplan⁵ predicted the existence of a new class of nonlinear optical effects involving the abrupt switching between the total-internal-reflection (TIR) and the normal-reflection (NR) regimes when the intensity of an incident beam or its angle of incidence was varied. In that paper, Kaplan considered the interface between a linear and a nonlinear Kerr medium as a remarkably simple system in which the effects could be identified. Since then, the nonlinear interface has been studied extensively, particularly by Smith, et al at Bell Laboratories⁶. The behavior

5. A.E. Kaplan, JETP Lett. 24, 114 (1976).

6. P.W. Smith, W.J. Tomlinson, P.J. Maloney and J.P. Hermann, IEEE J. Quant. Electron. QE17, 340 (1981).

of a nonlinear interface has turned out to be somewhat more complicated than originally anticipated: in particular, the very existence of optical bistability appears to be rather controversial.

From a theoretical viewpoint, the difficulty seems to be centered on the fact that plane waves are not stable solutions of Maxwell equations in a nonlinear medium. Attempts at a more realistic formulation of the problem, unfortunately, tend to obscure the elegant simplicity of the original results. Clear-cut experimental results, on the other hand, cannot be produced easily because of the difficult requirements of grazing incidence and high incident power.

In order to allow a more relaxed experimental setting, we have investigated a nonlinear frustrated total reflection cavity as an alternative system in which to study the Kaplan effect; in fact, this system, though much more complex than the nonlinear interface, should allow the choice of a more convenient angle of incidence and lower incident power. A schematic diagram of the system is shown in Figure 1.

Two well-known features of this device in the linear regime of operation (low power levels) are:

i) *The angular reflectivity spectrum is characterized by deep and sharp minima whose angular position is related to the parameters of the structure (thickness and optical constants); and*

ii) *At resonance, a very strong electric field builds up in the cavity layer with enhancements of two to three orders of magnitude with respect to the intensity of the incident beam.*

When either the cavity layer, or the substrate is made of a nonlinear material (e.g., a positive Kerr medium) the index of refraction can be made to suffer significant variations which result in nearly discontinuous reflectivity changes and in the occurrence of hysteresis.

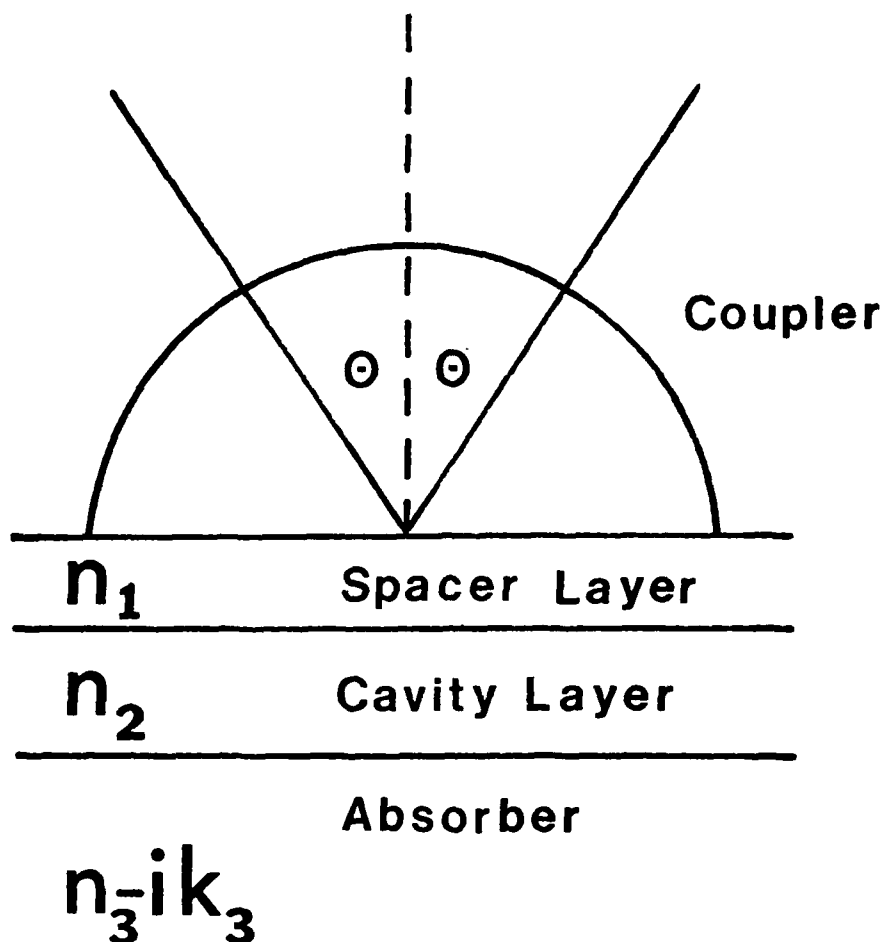


Fig. 1 Schematic configuration of a Frustrated Total Internal Reflection Cavity

This type of behavior suggests the possibility of using such a device as a switch for the protection of sensitive optical components. At low incident power levels, the structure would provide a high reflectivity and a detector could be safely exposed to the reflected radiation. Above a certain threshold level, which presumably could be adjusted to be low enough not to damage a sensor, the reflectivity would drop to very low values, thus protecting the sensitive surface.

Additional information can be found in Paper 3 listed at the end of this report.

3) Reviews of past work

Two review papers were delivered as invited talks at the *Synergetics Conference* in Berlin, and at the *NATO Summer School on Nonequilibrium Cooperative Phenomena*. These papers listed as numbers 4 and 5 at the end of this report contain information on the single and multi-mode operation of a bistable system, on hybrid bistability with delay and on the laser with injected signal. Paper 5, in particular, contains an extensive tutorial section outlining the basic principles of operation of a bistable device and comments on the practical applications. Earlier work by members of our group is described in a review paper entitled "*Absorptive Optical Bistability and Self-Pulsing*" which was requested by the Chinese review journal, *PROGRESS IN PHYSICS*. This article (6 on the list) has appeared only in its Chinese translation.

2. THE LASER WITH INJECTED SIGNAL

The suggestion that a number of nominally identical lasers could be driven synchronously by a common master oscillator in a phase-locked configuration dates as far back as the early 1960's. Injection locking since then has been the subject of active consideration because of its interesting engineering applications. We were, apparently, among the first to carry out a detailed analysis of the behavior of a coherently driven laser¹ and of its stability properties. although limiting cases where the atomic variables are eliminated adiabatically and external field² or pump modulation³ are included, should also be noted. Over the past two years, we have analyzed the more interesting situation where the injection and pump parameters are constant in time, and all the instabilities are self-induced within the system.

Depending on the gain of the laser and the setting of the various resonances, the driven laser develops a pulsed output over a range of driving field strengths; eventually, above a certain injection locking threshold, it becomes stable again with its output being phase locked with the driving field. The instability region displays a remarkable variety of pulsing conditions. These have been described for a very high gain system in Paper 7. They include regular intensity modulation at low driving fields which are followed at higher intensities by irregular pulsations, bursting action, reverse period doubling bifurcations, breathing patterns and spiking.

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1. L.A. Lugiato, Lett. Nuovo Cimento 23, 609 (1978) and referenced cited therein.
 2. T. Yamada and R. Graham, Phys. Lett. 53A, 77 (1975).
 3. M.J. Scholz, T. Yamada, H. Brand and R. Graham, Phys. Lett. 82A, 321 (1981).

This remarkable sequence is illustrated in *Figures 2 and 3*. In this paper we also give convincing evidence for the existence of a strange attractor by showing the exponential divergence of nearby trajectories in the chaotic regime (*Fig. 4*). We have also carried out a detailed investigation of a driven laser with a lower gain setting and revealed the existence of co-existing basins of attraction and hysteretic effects. Our understanding of the mechanism that governs the emergence of erratic behavior is still primitive, but with the combined analysis of the output power spectra, Poincaré maps and the entire set of Lyapunov exponents we have produced a detailed picture of bifurcation set over most of the range of interest of the driving field.

(*Papers 8 and 9, listed at the end of this report.*)

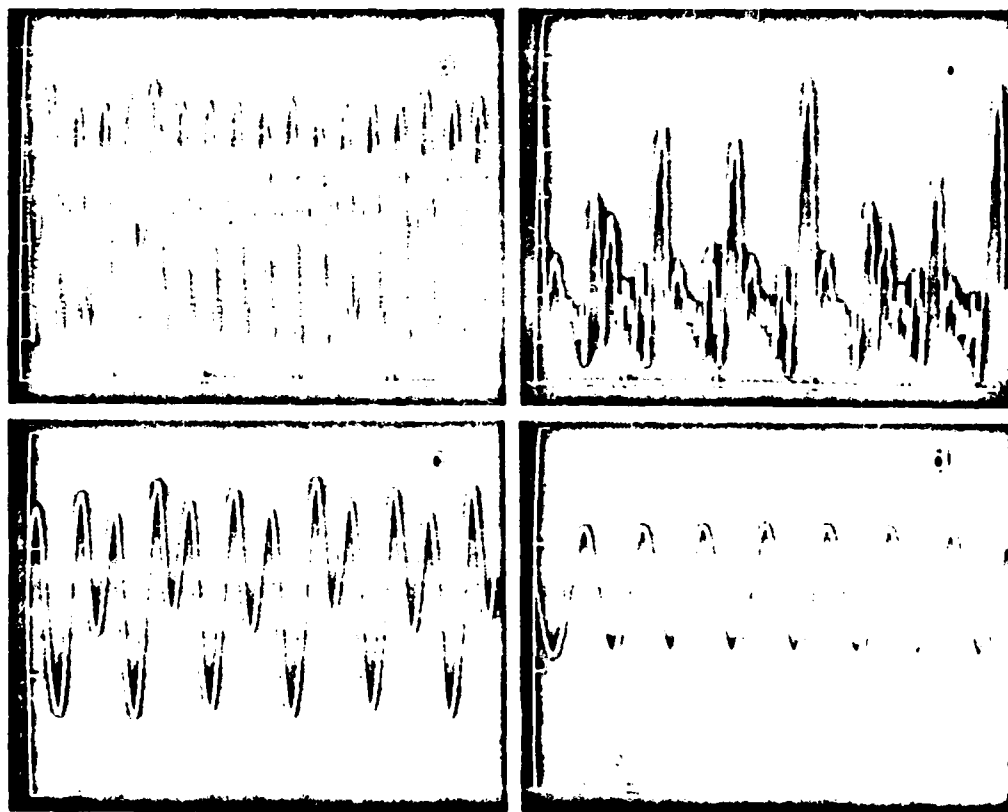


Fig. 2. Time evolution of the normalized emitted field $|x|$ for $C = 500$, $\Delta = \theta = 5$, $\tilde{\kappa} = 1$, and $\tilde{\gamma} = 1$. The horizontal axis is measured in τ units: (a) erratic behavior, $\gamma = 117$; (b) bursting, $\gamma = 250$; (c) 4P-type solution, $\gamma = 279$; (d) 1P-type solution, $\gamma = 300$.

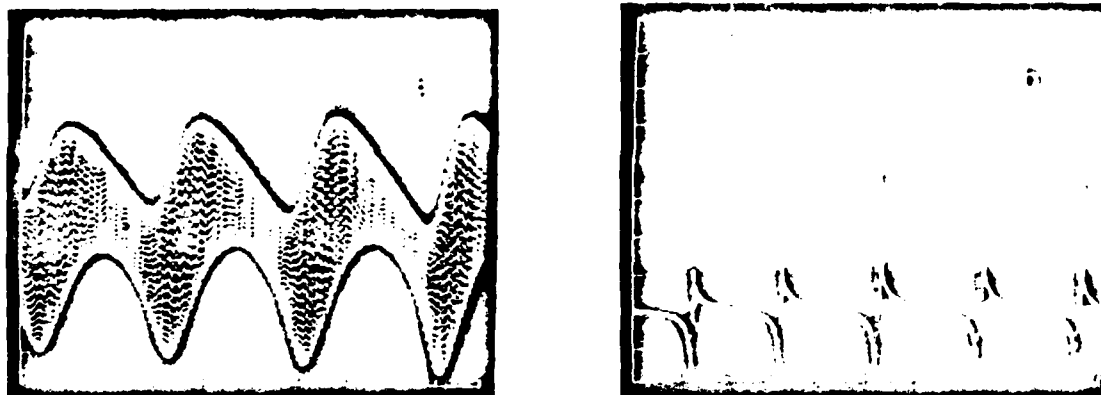


Fig. 3. Time evolution of the normalized emitted field $|x|$ with the same operating parameters as in fig. 2. For the chosen values of the driving field, the system displays: (a) a marked modulation of the self-pulsing envelope (heavy breathing), $\gamma = 310.3$; (b) spiking action, $\gamma = 311$.

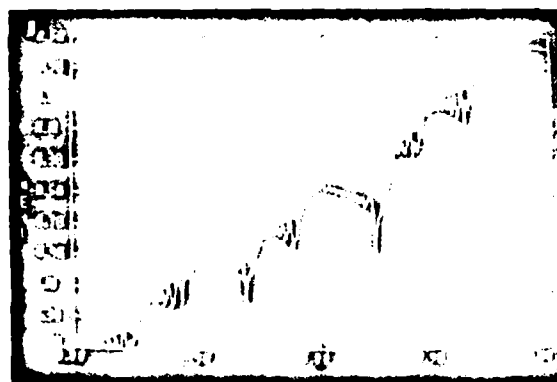


Fig. 4. The logarithm of the cartesian distance between two initially "close" trajectories grows, on the average, linearly with time. This plot shows evidence of "exponential divergence" when a trajectory evolves on the domain of the strange attractor.

We believe this latter publication to be significant because it demonstrates, with a specific nontrivial example, the value of these combined techniques for the analysis of complicated bifurcation patterns. We have also made an effort to explain in layman's terms, how the Lyapunov exponents are defined, what their main properties are, and which signatures are to be expected in connection with specific bifurcations.

3. TRANSVERSE EFFECTS AND INSTABILITIES

With very few exceptions¹⁻⁵, most instability problems in quantum optics have been analyzed in the context of the uniform plane wave approximation, an idealized situation where the active or passive atoms are immersed in a field of equal strength regardless of their distance from the axis of the system. In general, this approximation is regarded as a reasonable first step and most features are expected to be qualitatively correct, even in the presence of a nonuniform intensity profile. Actually, some evidence already exists that transverse beam nonuniformities can alter some prediction of the plane wave theory, not only quantitatively, but qualitatively as well. Thus, for example, the Ikeda instability which in the plane wave approximation is the source of a period doubling sequence and chaotic oscillations, develops a different route to chaos if the input field has a Gaussian transverse profile². Other instabilities of the plane wave theory are even suppressed altogether in the presence of a Gaussian field profile.

Thus, the inclusion of a radially nonuniform cavity field is not only a step forward from the viewpoint of realistic modelling, but may also be necessary for a detailed comparison of the theoretical predictions with experiments. As one may easily appreciate such extensions of the plane wave theory, it is not an easy matter from a technical point of view. As a first step, we have considered a situation where a cavity resonator can

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1. R.J. Ballagh, J. Cooper, M.W. Hamilton, W.J. Sandle, D.M. Warrington, *Opt. Comm.* 37, 143 (1981).
 2. J.V. Moloney, F.A. Hopf and H.M. Gibbs, *Phys. Rev.* A25, 3442 (1982).
 3. W.J. Firth, E.M. Wright, *Phys. Lett.* 92, 211 (1982).
 4. L.A. Lugiato, M. Milani, *Zeit. Phys.* B50, 171 (1983); *Opt. Comm.* 46, 57 (1983).
 5. R. Hauck, F. Hollinger, H. Weber, *Opt. Comm.* 47, 141 (1983).

support only a TEM_{00} mode whose radial profile is Gaussian. If an input field is present, we assume that this is matched to the fundamental mode of the resonator (an arrangement, for example, that has been realized by Kimble and collaborators at the University of Texas in Austin).

Our analysis of the effects of the radial profile on the operation of a bistable system, a free running laser, and a laser with an injected signal has been carried out in the mean field limit and for a cavity arrangement corresponding to a large Fresnel number, so that the beam radius is practically constant over the longitudinal dimension of the atomic sample.

We have concentrated our attention on instabilities which are known to occur in the plane wave limit and we have evaluated the role played by the more realistic field profile. We expect our one-transverse-mode assumption to be satisfactory in at least two situations: (i) *when all the other modes are sufficiently detuned from the carrier frequency of the incident field (or, in the case of the free running laser, from the center of the gain profile)* or (ii) *when the losses of all the other modes are large enough to keep their amplitudes at a negligible level.* In the absence of arguments for neglecting higher order transverse modes, a compelling justification for the stability analysis performed in this work is that our model allows an analytic handling of the steady state stability properties over the entire control parameter space. The exact equations of motion without the one-mode assumption can be studied, apparently, only by numerical methods--a task which is both time-consuming and of uncertain effectiveness. Thus, only as a subsequent step, and building from the knowledge gained from the present model, do we plan to study a more general setting. A future extension of this work will also involve the inclusion of inhomogeneous

atomic broadening.

An important result of our study is the identification of the optimum parameter setting for the observation of unstable behavior. We have predicted, for example, that cavity and atomic detunings with opposite signs favor the emergence of unstable behavior. This fact has already been confirmed qualitatively in a very recent observation of self-pulsing in single-mode optical bistability by Kimble and collaborators. In addition, we have discovered an enhancement of the domain of instability in correspondence with the disappearance of a hysteresis region in the state equation of both bistability and laser with injected signal.

A surprising result is the disappearance of the self-pulsing instability, predicted to occur in a plane wave homogeneously broadened laser, when the cavity beam acquires a radial Gaussian profile. We cannot exclude that this instability will re-emerge for different values of the control parameters. Still, our calculations suggest that the observation of this so-called second laser threshold will be even more difficult than already anticipated in the plane-wave limit. *Additional technical details can be found in Paper 10 listed at the end of this report.*

4. UNSTABLE BEHAVIOR OF INHOMOGENEOUSLY BROADENED HIGH GAIN LASERS

The departure away from a stationary state of a homogeneously broadened cw pumped laser and the emergence of time-dependent output oscillations has been a subject of active research as far back as the early 1960's^{1,2}. Idiatulin and Uspenskii³, in a surprising analysis, showed that even a small amount of inhomogeneous broadening in the active medium is sufficient to produce significant qualitative changes in the behavior of the laser; in a laser medium comprised of two groups of atoms with different resonant frequencies, they discovered, for example, a significantly lower threshold for instability than with the corresponding homogeneous system. Lower thresholds for unstable behavior in the presence of inhomogeneous broadening have been a confirmed feature of all studies up to the present time. It is then of great interest, as well as a matter of practical importance, to investigate the operation of inhomogeneously broadened lasers, since lasers of this type comprise the majority of practical laboratory devices. In fact, by far, the most common types of gain profiles in laser media are broadened by atomic thermal motion in gaseous systems, or by local field perturbations in solid state materials. At low gas pressure, of course, thermal motion is responsible for Gaussian

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1. A.Z. Grazyuk and A.N. Oraevskii, Radiotekh. Elektron. 9, 524 (1964) [Radio. Eng. Electron. Phys. (USSR) 9, 424 (1964)]; and Quantum Electronics & Coherent Light, ed. P.A. Miles (Acad. Press: NY (1964)) p. 192.
 2. H. Haken, Z. Phys. 190, 327 (1966); H. Risken, C. Schmid, W. Weidlich, Z. Phys. 194, 337 (1966); R. Graham, H. Haken, Z. Phys. 213, 420 (1968); H. Risken, K. Nummedal, J. Appl. Phys. 39, 4062 (1968).
 3. V.S. Idiatulin, A.V. Uspenskii, Radio. Eng. Electron. Phys. (USSR), 18, 422 (1973).

broadening; lineshapes in solids are typically more complicated with their most prominent features resembling, sometimes, suitably broadened Lorentzian lines.

Casperson⁴ demonstrated the occurrence of transitions from cw to pulsed operation in a broadened laser model by numerical integration of the equations of motion for a collection of four-level atoms in a single-mode, standing wave cavity. With the inclusion of a sufficient number of essential parameters, his numerical simulations produced results in substantial agreement with the measured threshold values and even with the output pulse shapes recorded in earlier experiments with a 3.51 μm high-gain xenon laser. More recent experiments by Abraham and collaborators⁵ have probed these instabilities in greater detail using lasers similar to Casperson's, in addition to unidirectional xenon ring lasers and to standing wave systems operating at the 3.39 μm transition of neon.

Recently we demonstrated the existence of multiple steady state solutions for a sufficiently detuned laser with a Lorentzian broadened lineshape and raised the possibility that bistable behavior and hysteresis may be observed between zero and nonzero intensity branches (*Papers 11,12*). This would manifest itself as a highly unusual sudden turn-on of the laser intensity at the first threshold. Additional work on the stability properties of Lorentzian and Gaussian lasers has also been carried out by one of our European collaborators, Paul Mandel⁶ and by Hendow and Sargent⁷.

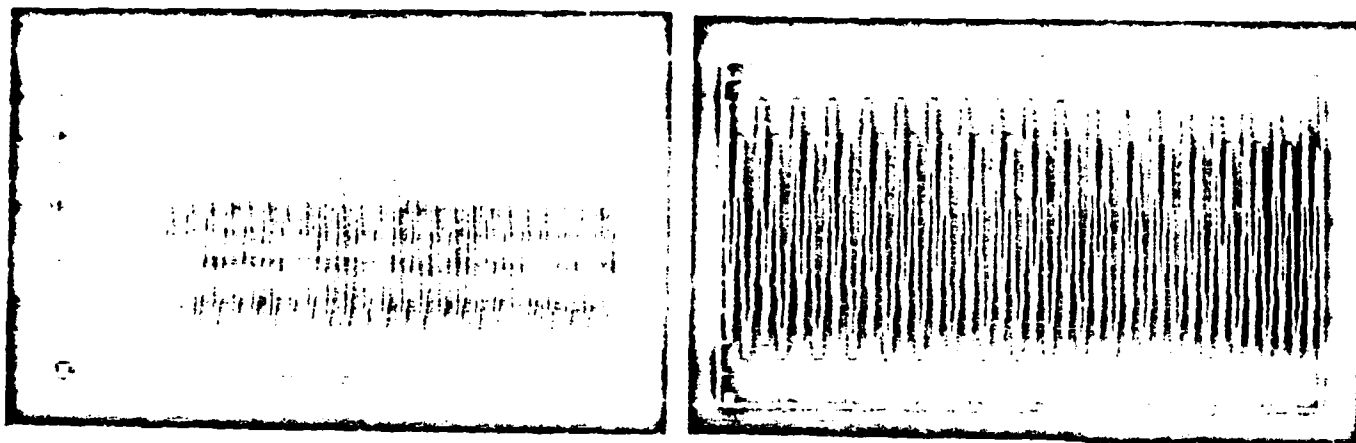
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4. L.W. Casperson, *IEEE J. Quant. Electron.* QE14, 756 (1978); and *Lecture Notes in Physics*, 182, 88 (1983), (Springer Heidelberg).
 5. N.B. Abraham et al., *Lecture Notes in Physics*, 182, 107 (1983) Springer Heidelberg).
 6. P. Mandel, *Opt. Comm.* 44, 400 (1983); *Opt. Comm.* 44, 269 (1983).
 7. S.T. Hendow, M. Sargent III, *Opt. Comm.* 40, 385 (1982); *Opt. Comm.* 43, 59 (1982).

In a joint collaborative effort with N. Abraham, D. Bandy, L. Lugiato, P. Mandel, we have recently developed the appropriate formal tools for the analysis of the steady state and stability properties of an inhomogeneously broadened laser with an arbitrary lineshape. In a paper which has been submitted for a special issue of the *JOURNAL OF THE OPTICAL SOCIETY*, we have provided a detailed comparison between Gaussian and Lorentzian broadening, including a study of the steady state and of the stability of the linearized equations of motion.

We have used our detailed knowledge of the domains of instability in the space of the control parameters to investigate the temporal evolution of the output intensity. This problem presents the usual challenges of any fairly large-scale numerical simulation. The continuous atomic profile must be suitably discretized into a number of components, or "packets", so that the number of equations to be solved simultaneously is usually rather large (typically about 300 in most of our simulations).

As expected on the basis of the linear stability analysis, self-pulsing behavior is a frequent occurrence even at low gain levels. Examples of oscillations displaying period doubling for measuring values of the gain are shown in *Figs. 5a,b*.

A very interesting result of our simulation is the appearance of trains of pulses when the population relaxation rate becomes sufficiently smaller than that of the polarization. The observed pulses--an example of which is shown in *Fig. 6*--have a peak intensity that appears to scale as the square of the atomic inversion density. This feature of the solution, coupled to the presence of ringing in the tail of the pulses and to the considerable delay (lethargy) between successive peaks is highly suggestive of the presence of cooperative behavior. We have no proof of this suggestion at the present time, but work is underway to clarify this issue.



Figs 5a and b An example of period doubling bifurcation in a Free Running Laser with inhomogeneous broadening

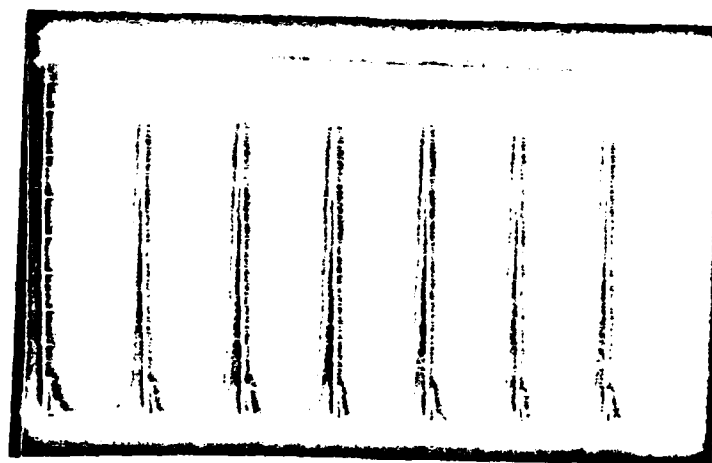


Fig. 6 Spiking action in a Free Running Laser with inhomogeneous broadening

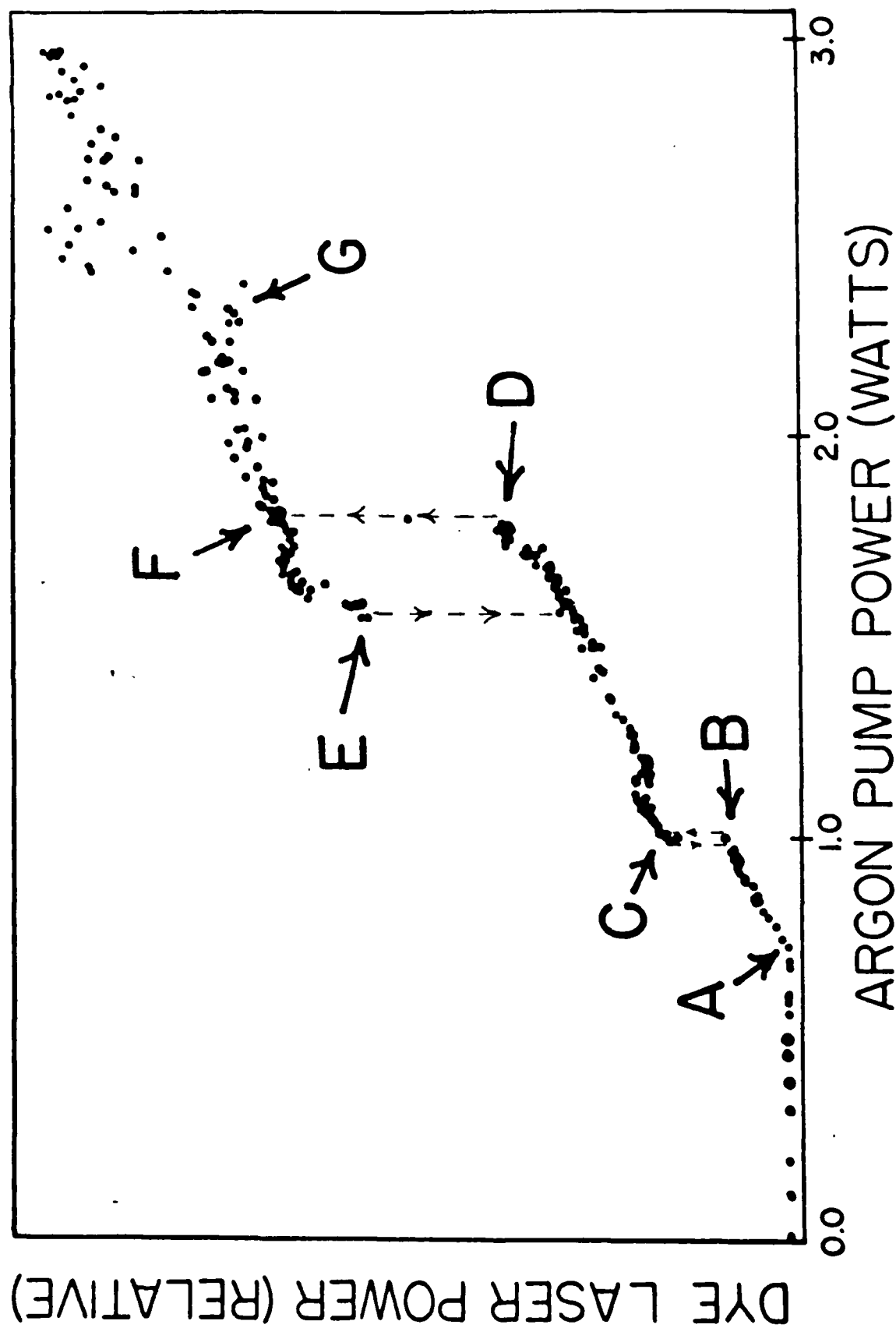


Fig. 7 Output intensity of a multi-mode homogeneously broadened laser as a function of the pump power. These results have appeared in L.W. Hillman et al., Phys. Rev. Lett., 52, 1605 (1984).

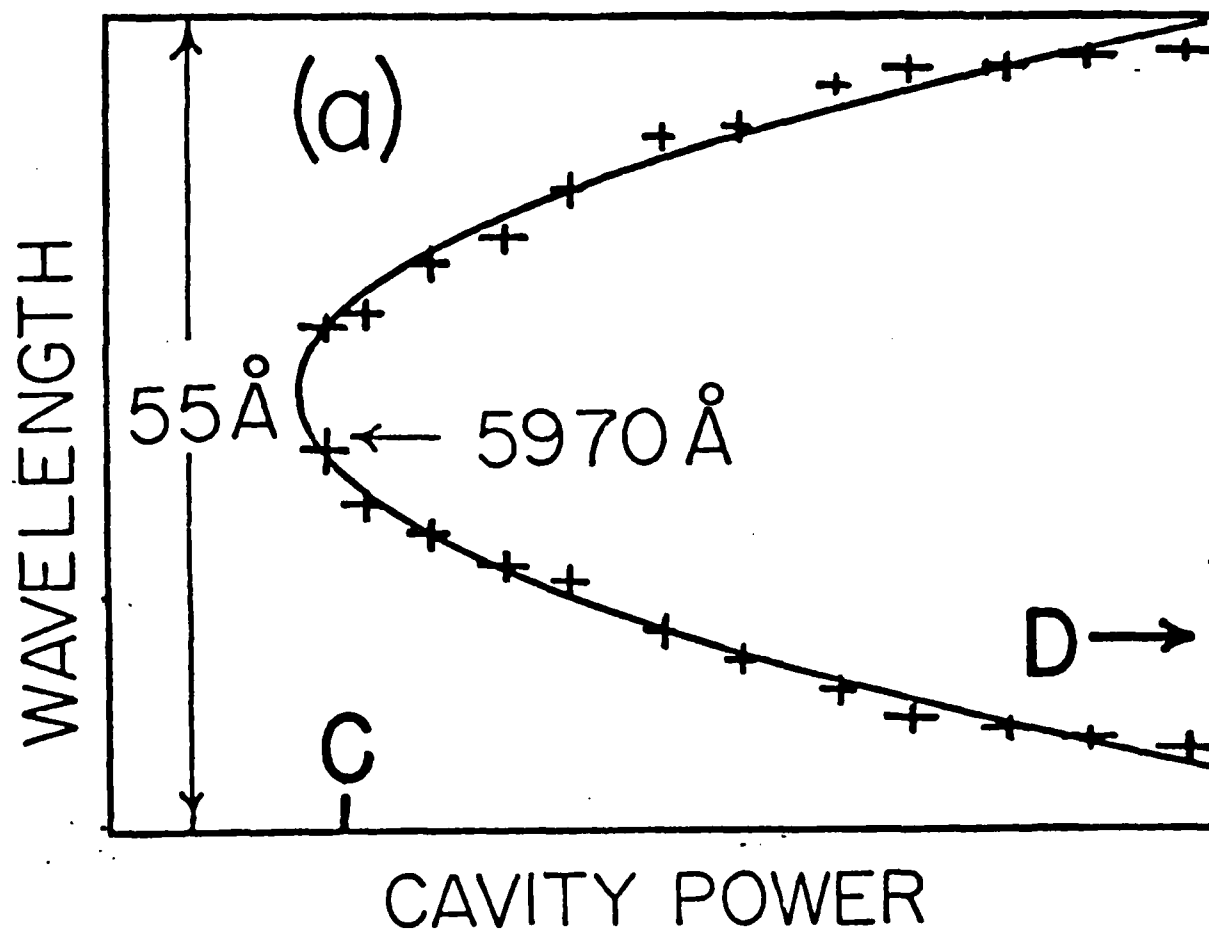


Fig. 8 Sideband splitting as a function of the pump power in a multimode homogeneously broadened laser. These results have appeared in L.W. Hillman et al., Phys. Rev. Lett., 52, 1605 (1984).

5. MULTI-MODE INSTABILITIES IN HOMOGENEOUSLY BROADENED LASERS

In an important recent contribution¹ Hillman and collaborators demonstrated for the first time the existence of new stable operating states of a homogeneously broadened laser consisting of a simple ring cavity with no modulators or saturable absorbers. As shown in Figs. 7,8 of Ref. 1, the output intensity of the laser undergoes remarkable variations as a function of the power of the driving pump laser. Point A marks the usual, so-called first, laser threshold where the incoherent fluorescent intensity becomes a linear function of the atomic inversion (or pump power) while the laser linewidth undergoes the well known narrowing that accompanies the crossing of the threshold. At point B, the output intensity suffers a discontinuous jump to a higher value (point C) whereupon it grows again linearly with the pump power. At point D, a second discontinuous jump is clearly observed to a higher, noisier branch. On reversing the direction of variation of the pump laser power, downward jumps are observed with clear hysteresis effects.

These discontinuous transitions are already matters of great interest because they are entirely unexpected in the single-mode operation of a homogeneously broadened laser; they provide direct evidence for the important role played by the nonresonant cavity modes. Furthermore, the spectrum of the laser output at point B is characterized by the sudden disappearance of the single resonant line and the appearance in its place of two split sidebands whose spacing grows linearly with the Rabi frequency of the

1. L.W. Hillman, J. Krasinski, R.W. Boyd, C.R. Stroud, Jr., Phys. Rev. Lett. 52, 1605 (1984).

circulating laser field. The experimental evidence points to a mode of operation in which sidebands have become unstable and have quenched the resonant mode. A linear stability analysis of a multi-mode homogeneous laser² is in agreement with the existence of unstable sidebands; however, a more detailed analysis of ours, based on a three-mode model under the adiabatic scan conditions of the Hillman experiment, is presently unable to account for the sudden intensity switching in the absence of a substantial amount of noise.

In collaboration with P. Mandel and L.A. Lugiato, we have also analyzed the time-dependence of the three-mode model and confirmed that, even in this case, solutions with zero intensity for the resonant mode and nonzero sidebands exist, but require a sizeable initial fluctuation in order to be excited (hard-mode instability). The experiment by Hillman and collaborators represents an important technical advance, because until now, the only instabilities that have been well documented experimentally in active media have involved inhomogeneously broadened lasers and lasers with a saturable absorber. Theoretical efforts must be developed to clarify the present apparent discrepancies between our linear stability analysis and time-dependent solutions and the observed behavior of the laser output intensity.

2. H. Risken, K. Nummedal, J. Appl. Phys. 39, 4062 (1968).

6. LOGISTIC MAPS AS DISCRETE MODELS OF COUPLED DYNAMICAL SYSTEMS

Period doubling bifurcations leading to chaos, best illustrated by the logistic equation $x_{n+1} = 4\lambda x_n (1-x_n)$, have been observed experimentally and theoretically in many dynamical systems. There exists, however, physical problems, such as, AC-driven DC superconducting quantum interference devices (SQUID), energy transfer between two bond modes of a polyatomic molecule and population competition between biological species, which can best be described by the coupling of two subsystems, each of which alone can undergo period doublings. In order to understand the global dynamics of such coupled systems, for instance, as a function of nonlinearity and coupling strength, we have studied a general model based on the coupled logistic maps.

$$\begin{aligned}x_{n+1} &= 4\lambda_1 x_n (1-x_n) + G_1(x_n, y_n) \\ y_{n+1} &= 4\lambda_2 y_n (1-y_n) + G_2(x_n, y_n),\end{aligned}$$

where G_1, G_2 are coupling terms. We have focused our attention on the following two cases: (i) *symmetric bilinear coupling* $G_1 = G_2 = \gamma x_n y_n$ and (ii) *linear coupling* $G_1 = \gamma y, G_2 = \gamma x$.

A numerical search in the three-dimensional control parameter space leads to the following results:

- a) If we fix the value of γ , and move on the (λ_1, λ_2) plane of the control parameter space along the $(\lambda_1 = \lambda_2)$ diagonal with increasing γ , chaotic behavior appears via complicated dynamical behavior, including Hopf bifurcations, quasiperiodic and phase-locking, etc. A detailed bifurcation scheme in the case $\gamma = 0.1$ will be reviewed below.

- b) Away from the diagonal region, chaos may arise as the result of an infinite sequence of period doubling bifurcations.
- c) The diagonal region of the (λ_1, λ_2) plane that displays quasiperiodic behavior leading to chaos grows in size as the coupling strength γ increases.

In order to reveal the detailed mechanisms to chaos around the diagonal region, we have carried out an extensive study of the case $\gamma = 0.1$ and $\lambda_1 = \lambda_2$ by using the Lyapunov spectral method. The results are summarized in Fig. 9, where the symbols are explained in the figure caption. In addition to the domains of periodic, quasiperiodic and chaotic behavior, we have identified nine cases of crises (sudden changes of chaotic attractors). Five of them belong to a type called "boundary crisis" and are denoted by arrows in the figure. The other four, denoted by the endpoints of double horizontal lines, belong to a new type of crisis called "cyclic crisis". In such a crisis, several chaotic attractors undergo cyclic collisions with their basin boundaries and merge into a bigger one, in which chaotic phase trajectories exhibit cyclic transitions among the original components with a well-defined overall period.

Additional technical details can be found in Papers 13 and 14 listed at the end of this report.

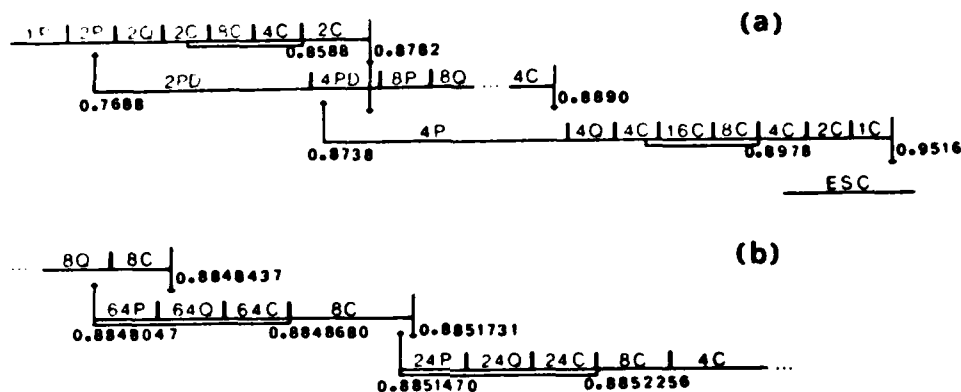


FIG. 9 Bifurcation scheme of the map (1) with $\gamma = 0.1$. The attractors are denoted by P (periodic), PD (periodic attractor located on the diagonal), Q (quasiperiodic), C (chaotic), and ESC (escape to infinity). The integer n that precedes the symbols indicates that the attractor can be divided at most into n separate parts such that each part is an attractor of the map $F^{(n)}$ ($F^{(n)}(x, y) \equiv F(F^{(n-1)}(x, y), F^{(n-1)}(y, x))$, $F^{(1)} \equiv F$). We use multiple horizontal lines to represent several coexisting attractors with their own basins. Double lines under an attractor symbol denote coexisting attractors which are mirror images of each other. Frequency locking and windows within the quasiperiodic and chaotic regions are not indicated.

7. COLLECTIVE INSTABILITIES AND HIGH-GAIN REGIME IN A FREE ELECTRON LASER

Previous studies of the free electron laser (FEL) in the high gain regime¹⁻³ have shown that with an appropriate selection of the electron bunching can undergo exponential growth as a result of a collective instability of the electron beam-undulator-radiation field system. In this project, we focused on the conditions for the onset of this instability using a new secular equation for the characteristic complex frequencies of the FEL system. On the basis of these results, we have shown how one can rederive the small-signal gain formula and established the conditions for its validity. We also considered the problem of the initiation of laser action and of the growth of the radiation field from noise, and proposed a formula to evaluate the lethargy (build-up) time of the first pulse. Finally, we analyzed in detail the nonlinear regime of the FEL by numerical methods and obtained results that suggested the existence of an optimum efficiency of the device.

In the derivation of our working equations, we selected the phase and the energy as the basic electron variables, and assumed the slowly varying phase and amplitude approximation for the radiation field, as done in earlier developments^{4,5}. Unlike earlier works, on the other hand, we have

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1. N.M. Kroll and W.A. McMullin, *Phys. Rev.* **A17**, 300 (1978).
 2. G. Dattoli, A. Marino, A. Renieri, F. Romanelli, *IEEE J. Quant. Electron.* **QE17**, 1371 (1981).
 3. R. Bonifacio, F. Casagrande, G. Casati, *Opt. Comm.* **40**, 219 (1982).
 4. W.B. Colson, S.K. Ride, in *Physics of Quantum Electronics*, edited by S.F. Jacobs et al. (Addison-Wesley: MA, 1980) Vol. 7, p. 377.
 5. C. Pellegrini, in *Free Electron Lasers*, edited by A.N. Chester and S. Martellucci (Plenum Press; NY (1983)).

not restricted the electron bunch to retain nearly its initial speed during the generation of the FEL signal. Thus, no limitation is imposed ab-initio on the available laser gain. Our new instability condition represents an extension and generalization of the one proposed by Bonifacio and collaborators in *Reference 3*. Below threshold for laser amplification, the output field displays only small amplitude oscillations when plotted as a function of time (see Figure 10 below) which we have been able to reproduce analytically



Fig. 10

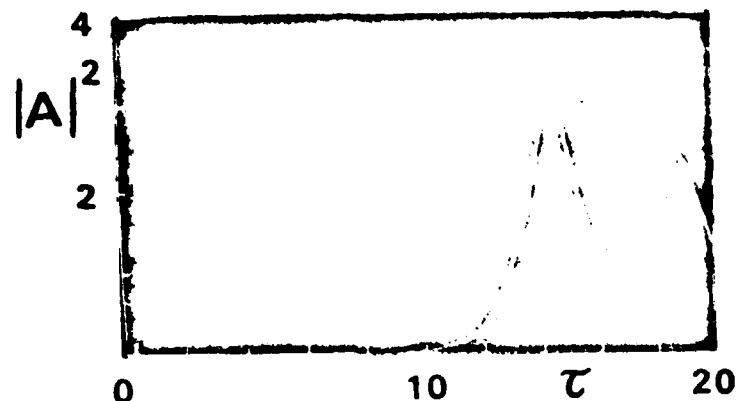
Output intensity $|A|^2$ for $\rho = 0.01$ and $\delta = 4.0$. The eigenvalues of the linearized equations are $-0.519, 0.628, 3.066$. The modulation is due to the beat of the different exponential terms in the solution.

with great accuracy. The oscillations can easily be understood as beat patterns among the frequencies of the linearized modes of the problem.

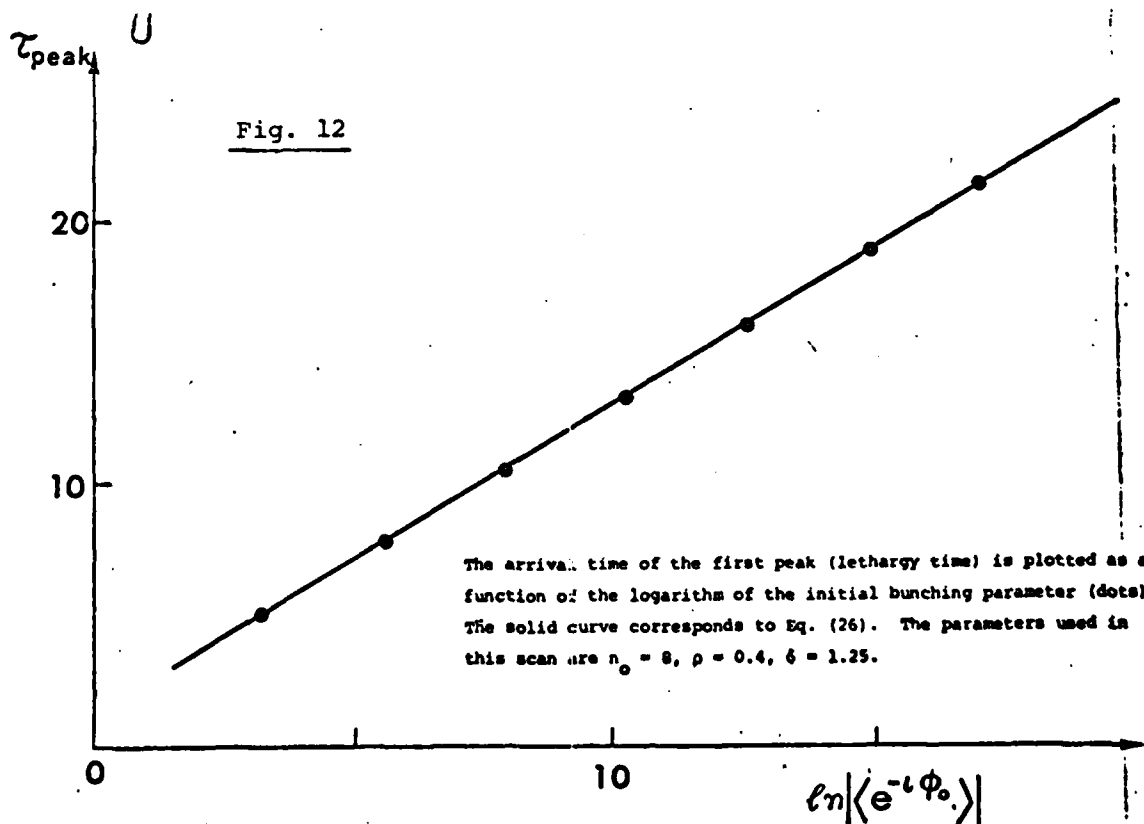
The evolution above threshold for laser amplification is entirely different, as shown in *Fig. 11*, for the case of an initial zero field and a small initial bunching parameter (this is the analogue of the macroscopic polarization in an ordinary laser problem). Under unstable conditions, fluctuations in the electron injection velocities, or the lack of uniformity in the initial distribution of the electron phase variables, or the presence of an initial field, will trigger the growth of the signal. This behavior is very general, and is independent of the details of the initial triggering mechanism as long as this perturbation is small. In our *Paper 15*, we have reported a semi-empirical formula describing the time of arrival of the first peak of the FEL pulse. This quantity is an essential design parameter that translates into the required undulator length according to the formula $L = c \tau_{\text{peak}}$ where c is the speed of light. The agreement between our predicted peak arrival time and the calculated time from the numerical simulations for different initial values of the bunching parameter is very impressive (see *Fig. 12*).

Additional technical details can be found in Paper 15 listed at the end of this report.

Fig. 11



Output intensity $|A|^2$ versus time above threshold. The parameters used in this simulation are $\rho = 0.0021$, $\delta = 1.86$, $n_0 = 16$.



8. BISTABILITY AND HYSTERESIS IN LASER-DRIVEN POLYATOMIC MOLECULES

It is well known that classical solutions of damped and driven anharmonic oscillators show bistable behavior, in the sense that the asymptotic amplitudes of oscillation can approach two different stable values over a certain domain in the control parameter space of the driving frequency and amplitude. In fact, the steady state amplitude, plotted as a function of the driving frequency and amplitude of the forcing term, forms a surface with the shape of a cusp catastrophe. The middle portion is unstable, while the other two sheets are stable in most cases. Huberman and Crutchfield¹ have shown, however, that the upper branch of a Duffing oscillator can become unstable, and that this instability leads to an infinite sequence of period doubling bifurcations and chaos.

We have made an attempt to investigate the unstable properties of a damped and driven quantum anharmonic oscillator developed several years ago by Narducci and collaborators in connection with the multi-photon absorption of polyatomic molecules². We found that the steady state surface, as in the case of a classical anharmonic oscillator, takes the shape of a cusp catastrophe³. A linear stability analysis shows that the upper branch is always unstable. Depending on the control parameters of the system, part of the lower branch may become unstable and develop Hopf bifurcations with unstable limit cycles. In physical terms, our results imply that

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1. B.A. Huberman, J.P. Crutchfield, *Phys. Rev. Lett.* **43**, 1743 (1979).
 2. L.M. Narducci, S.S. Mitra, R.A. Shatas, C.A. Coulter, *Phys. Rev. A* **16**, 247 (1977).
 3. J.M. Yuan, E. Liu, M.W. Tung, *J. Chem. Phys.* **79**, 5034 (1983).

there is a threshold field intensity above which an ensemble of molecules can be excited easily, as long as the field frequency is sufficiently red-shifted from the fundamental frequency of the pumped mode. On the other hand, when the intensity is below this threshold value, there is a minimum average vibrational excitation such that an ensemble of molecules can still be effectively excited by a relatively weak field, as long as it is initially prepared with a sufficiently high vibrational excitation. A calculation using parameters that are appropriate to SF_6 molecules shows that its bistable region overlaps with that leading to multiphoton dissociation. Molecules with two or three atoms appear to be better candidates for experiments directed at showing bistable response.

Additional technical information can be found in Papers 16 and 17 listed at the end of this report.

9. TWO-PHOTON LASER AMPLIFICATION

The suggestion that an inverted atomic population could be forced to decay by two-photon emission dates back to the early 1960's^{1,2}, but in spite of some theoretical progress³⁻⁵, the experimental search for a two-photon laser is still in its infancy. M.M.T. Loy⁶, in 1978, reported the first experimental observation of population inversion, two-photon gain and emission from a two-photon transition in ammonia. Because of intrinsic limitations with the energy stored in the excited medium, the NH_3 system cannot be viewed as a very promising prospect for achieving two-photon laser oscillation, although the adiabatic rapid passage adopted to achieve population inversion may be of value in future work.

A more recent paper by Nikolaus, et al.⁷ reported the observation of degenerate and nondegenerate two-photon emission in atomic Lithium vapor pumped by two counter-propagating nitrogen-pumped dye laser beams. However, this was followed by some controversy which has left the matter in need of further experimental studies.

We have directed a considerable amount of experimental effort to observing two-photon gain between the $8p^2P$ and the $3p^2P$ states of Sodium. The strategy adopted in our experiments was to create complete inversion between two states of the same parity by single-photon absorption from the

-
1. P.P. Sorokin and N. Braslau, IBM J. Res. Dev. 8, 177 (1964).
 2. A.M. Prokhorov, Science 149, 828 (1965).
 3. L.E. Estes, L.M. Narducci, B. Shammas, Lett. al Nuovo Cimento 1, 175 (1971).
 4. R.L. Carman, Phys. Rev. A12, 1048 (1975).
 5. L.M. Narducci, W.W. Eidson, P. Furcinitti and D.C. Eteson, Phys. Rev. A16, 1665 (1977).
 6. M.M.T. Loy, Phys. Rev. Lett. 41, 473 (1978).
 7. B. Nikolaus, D.Z. Zhang and P.E. Toschek, Phys. Rev. Lett. 47, 171 (1981).

ground state, and to search for an emission signal at the complementary wavelength of a strong injected wave. To avoid the possibility of spurious effects due to the coupling of the pump with the injected wave, the latter was delayed by 20-50 nsec., preventing any spatial overlap.

Sodium in a 30 cm long heat pipe oven with a diameter of 2.5 cm and in the presence of Argon buffer gas at a typical pressure of 1.3 torr, was pumped by a 10 nsec. long UV signal ($\lambda = 0.2544 \mu\text{m}$) in resonance with the 3S to 8P transition. This UV pulse was obtained by second harmonic conversion of the $0.5088 \mu\text{m}$ output of a tunable nitrogen-pumped dye laser, and had a typical power of about 500 W. The presence of inversion was monitored by observing the fluorescence signal from the D-lines of the Sodium vapor at 90° to the direction of the propagation of the pump beam. Estimates of the excited state population based on the pump power and line width, in addition to the appropriate atomic parameters indicate that 1-2% of the original atoms should be in the initial excited state. Under the conditions of this experiment, a complete initial population inversion was established between the two levels of interest because the 3P level is initially empty.

The main objective of this experiment was to produce a two-photon signal at a wavelength of $1.14 \mu\text{m}$ in the presence of a strong injected pulse tuned in the region $0.7365\text{-}0.3785 \mu\text{m}$. This IR pulse was produced by a tunable dye laser pumped by a 100 MW Q-switched ruby laser. Infrared pulses of 5-10 MW with a beam diameter of 3-5 mm have been produced in this manner. Accurate timing between the single-shot ruby laser and the nitrogen-pumped dye laser, operating at about 10 Hz, was obtained with the help of appropriate timing electronics and by taking advantage of the slight temperature dependence of the delay of the ruby pulse. By monitoring the operating temperature of

the ruby cooling system, the delay of the infrared input pulse relative to the UV pump could be regulated to within a few nanoseconds.

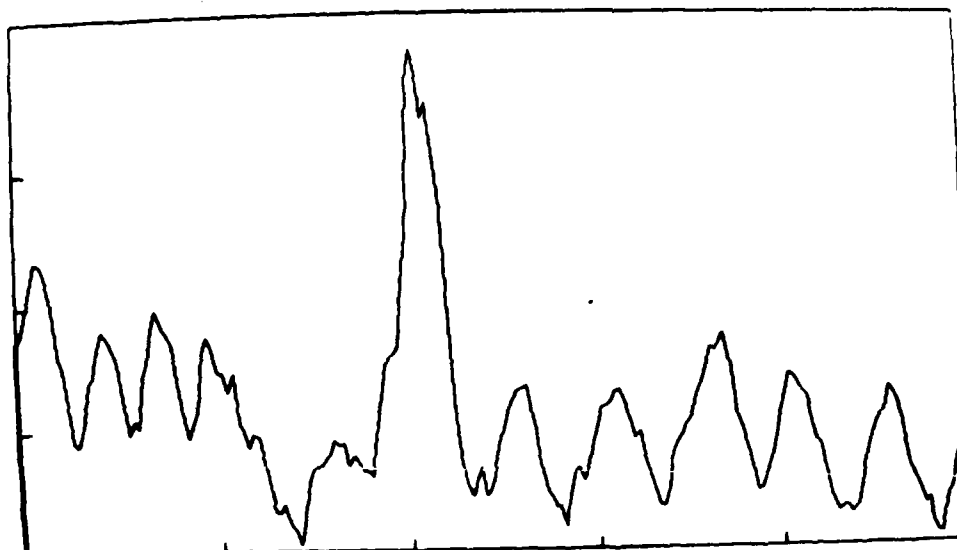
In the presence of an inhomogeneously broadened two-photon transition, the direction of highest gain for the $1.14\text{ }\mu\text{m}$ signal is expected to be opposite that of the strong input pulse because of partial cancellation of the Doppler broadening. Elimination of the almost ubiquitous input pulse was obtained with the help of antireflection coating on all the surfaces, and by placing appropriate filters and a small monochromator in front of the detector.

Due to the smallness of the expected signals, special care was taken to shield all connectors and cables from the RF noise of the nitrogen discharge. Some residual noise in the form of small amplitude oscillations was ascribed to interference from local radio transmitters.

An extensive search for reproducible signals with the required time delay from the pump pulse indicated that the optimum operating conditions require a relatively large vapor pressure of the Sodium gas (the outside temperature of the heat pipe oven was typically 500°C). Best results were obtained with about $6\text{ }\overset{\circ}{\text{A}}$ detuning of the infrared input pulse from the $8p^2P-4s^2S$ intermediate resonance. In spite of the smallness of the observed signals and the troublesome presence of residual RF noise, pulses at the required temporal position were observed consistently over several days, even after shutting the system off and turning it on several times. One of the characteristic features to be expected was the disappearance of the signals in the absence of the UV pump. This was observed with regularity by turning the pump signal on and off.

An example of the detected signal after the averaging of several shots is shown in *Figure 13*.

Additional information can be found in Papers 18 and 19 listed at the end of this report.



25 nsec / Division

Fig. 13 Two-Photon signal in Sodium after signal averaging

10. ADIABATIC ELIMINATION IN NONLINEAR DYNAMICAL SYSTEMS

The adiabatic elimination process is a procedure by which a number of dynamical variables can be suitably processed away from a set of coupled differential equations with the objective of reducing the description of a complicated system to a smaller number of degrees of freedom. As emphasized by Haken¹, the so-called adiabatic elimination process plays a fundamental role in understanding the origin of self-organization in open systems far from thermal equilibrium. As described in the context of the generalized Ginzburg-Landau equations for phase transition-like phenomena in open systems, this procedure is designed to cope with the behavior of nearly critical systems, a situation where the control parameters are adjusted in such a way that a stationary state is about to become unstable. In this case, part of the system, the so-called unstable modes, evolves through a very slow process of damping or amplification; thus, the remaining stable modes, which still evolve at their normal rates, can be eliminated adiabatically. With this setting, the accuracy of the procedure is controlled by the proximity of the system to the critical point and is normally adequate only in the local neighborhood of the transition.

In practice, the adiabatic elimination is often carried out in a global sense, usually after assuming that the rate constants of the variables to be eliminated are sufficiently larger than the remaining ones. Assessing the validity of this step is a much more delicate matter.

1. H. Haken, *Synergetics - An Introduction* (Springer, Berlin (1977)).

In *Paper 20*, we have analyzed this problem in detail using as a guideline the one-mode homogeneously broadened laser model, with an injected signal and arbitrary population difference, for added flexibility. The main result of our study is a set of five conditions under which the adiabatic limit can be insured to acquire global validity: these conditions involve not only the relative magnitude of the time scales, but also the magnitude of all the control parameters, of the physical variables and of their fluctuations. Numerous additional results include the generalization of the dressed mode description of dynamical systems and the clarification of the link between the adiabatic elimination procedure and the multiple-time-scale approach.

For numerous additional details, the reader is referred to Paper 20 listed at the end of this report.

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7. L.A. Lugiato, L.M. Narducci, D.K. Bandy and C.A. Pennise, "Breathing, Spiking and Chaos in a Laser with Injected Signal", *Opt. Comm.* 46, 64 (1983).
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12. PRESENTATIONS & INVITED TALKS

1. L.M. Narducci, L.A. Lugiato, N. Abraham, D.K. Bandy, "*Instabilities in a Single-Mode Inhomogeneously Broadened Laser*", Ann. Mtg. of the Opt. Soc. of Am., 1983.
2. D.K. Bandy, L.A. Lugiato, L.M. Narducci, "*Self-Pulsing and Chaos in a Laser with Injected Signal*", Ann. Mtg. of the Opt. Soc. of Am., 1983.
3. L.M. Narducci, J.Y. Gao, J.M. Yuan, M.F. Squicciarini, "*Route to Chaos in a Bistable System with Delay*", Ann. Mtg. of the Opt. Soc. of Am. 1983.
4. Y. Gu, J.M. Yuan, M. Tung, D.H. Feng, L.M. Narducci, "*Bifurcation and Crises in Coupled Logistic Maps*", Spring Mtg. of the Am. Phys. Soc., 1984.
5. L.A. Lugiato, L.M. Narducci, "*Gaussian Mode Instabilities in Optical Bistability and Related Systems*", Symposium on Optical Bistability, London (1984).
6. "*Unstable and Chaotic Behavior of a Delayed Bistable System*", a colloquium presented at the University of Rochester, 1983.
7. "*Unstable and Chaotic Behavior of a Delayed Bistable Laser System*", a colloquium presented at the University of Maryland (1984).
8. "*Unstable and Chaotic Behavior of Bistable Optical Systems*", a colloquium presented at the Naval Air Development Center, Warminster, PA (1984).
9. R. Bonifacio, C. Pellegrini, L.M. Narducci, "*Collective Instabilities and High-Gain Regime Free Electron Lasers*", Topical Met. on Free Electron Generation of Extreme UV Coherent Radiation, Brookhaven Natl. Labs, NY (1983).
10. J.M. Yuan, L.M. Narducci, D.H. Feng, M.W. Tung, "*Instability and Chaotic Behavior of Two-Dimensional Quadratic Maps*", Spring Mtg. of the Am. Phys. Soc., 1983.

13. OTHER MAIN ACTIVITIES BY THE PRINCIPAL INVESTIGATOR

- Jointly with Professors N.B. Abraham and L.A. Lugiato, I have served as Co-Editor of a special issue of the *Journal of the Optical Society B*, devoted to Instabilities in Active Optical Media (January 1985).
- In collaboration with Professor N.B. Abraham and Dr. P. Mandel, I am preparing a review article for *Progress in Optics*, devoted to Pulsations in Laser Systems.
- I was the recipient of the 1984 Lindback Teaching Award.
- I have been serving as a consultant on Professor Lugiato's and Dr. Mandel's contracts with the *Scientific Commission of the European Communities*. The contract represents a multi-national effort by several leading European physicists to study the feasibility of designing an optical computer.

14. LIST OF PERSONNEL

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W.W. Eidson, Professor of Physics

J.M. Yuan, Associate Professor of Physics

D.K. Bandy, Post-Doctoral Research Associate
(Previously a Graduate Research Assistant during funding of this project.)

D.D. Hughes, Assistant to the Project Director

M. Squicciarini, Graduate Student

M.W. Tung, Graduate Student

W.P. Zhang, Graduate Student

Elia Eschenazi, Graduate Student

M. Rankin, Graduate Student

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15. DEGREES AWARDED WITH PARTIAL SUPPORT OF FUNDING

Donna K. Bandy, Ph.D., June 1984

Dissertation: *"Instabilities, Self-Pulsing, and Chaos in
Optical Bistability and Laser with Injected Signal"*

Christine A. Pennise, BS, June 1984

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