A major purpose of the Technical Information Center is to provide the broadest dissemination possible of information contained in DOE's Research and Development Reports to business, industry, the academic community, and federal, state and local governments.

Although a small portion of this report is not reproducible, it is being made available to expedite the availability of information on the research discussed herein.

1

LA-UR 89-857

Received by OSTI

APR 0 6 1989

LA-UR--89-857

DE89 009392

TITLE OPTICAL CHAOS

AUTHOR(S) Peter W. Milonni

SUBMITTED TO Proceedings of the International School of Lasers and Applications, USSR, 17-23 March 1989.

OPTICAL CHAOS

Peter W. Milonni
Theoretical Division
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Abstract

The theoretical and experimental status of chaos in nonlinear optics and laser physics will be reviewed. Attention will then be focused on the possibility of chaotic behavior in individual atoms and molecules driven by intense radiation fields.

I. Introduction

I will begin by recalling a story that has been used to describe the relation between scientists and certain agencies that fund scientific research. In one version of the story there is a church in need of repair, and the priest imposes a rule that anyone wishing to enter the church must first spend an hour helping with these repairs. One day a pious man appears and asks the priest if he might possibly enter the church without doing any work, as he wants to deliver a message to a friend inside. "No," says the priest, "I know your kind. What you really came here for is to pray, and I won't allow it!"

I do not know of any direct applications of chaos to laser technology, and admit at the outset that I have come only to pray. It is particularly inspiring to do so here, where the patron saints have revealed so much about the mysteries of chaos.

Chaotic behavior has in recent years been observed in many laser and nonlinear optical systems.[1] It could have been (and probably was!) observed for many years, but only in the last decade or so has chaos been a subject of careful investigation rather than a nuisance to be avoided. For instance, it has long been observed that instabilities can develop in lasers and optical parametric oscillators with feedback. On the theoretical side, we might note an early paper by Buley and Cummings[2] in which a numerical simulation of a single mode laser indicated the possibility of output consisting of "a series of almost random spikes."

What is chaos good for? I recently saw a television show which described how some cardiologists are using ideas of chaos and strange attractors in research on heart fibrillations. But as I indicated earlier, I am not aware of any applications in laser technology where chaos has actually been *utilized*. At the present time chaotic behavior in atomic, molecular, and optical (AMO) physics is being studied not for any important applications, but simply because it is interesting in its own right, and because by looking at interesting things we gain a deeper appreciation and understanding of the world in general, and AMO physics in particular.

2. What is Chaos?

There now seems to be a general consensus on the *definition* of chaos: Chaos means very sensitive dependence on initial conditions, i.e., at least one of the Lyapunov exponents of the system is positive. By computing the Lyapunov exponents of a system, therefore, one can say unambiguously whether its dynamics are chaotic.[3]

To illustrate the idea of "very sensitive dependence on initial conditions" associated with a positive Lyapunov exponent, let us briefly review some properties of the Bernoulli shift $x_{n+1} = 2x_n$ (mod 1), i.e.,

$$x_n = 2^n x_o \pmod 1 \tag{2.1}$$

This is admittedly a well-worn example by now, but it illustrates the point so nicely that I am willing, for the benefit of the beginner, to carry a few coals to Newcastle. We note the following: (1) The system (2.1) evolves chaotically, i.e., with very sensitive dependence on initial conditions. For if the "initial condition" x_o is changed to $x_o + \epsilon_o$, then x_n is changed by $2^n \epsilon_o = \epsilon_o e^{n\log 2}$. It is worth pointing out here that "very sensitive dependence on initial conditions" in the definition of chaos means in fact *exponential* sensitivity, as in this example. In this example the number log2 is the Lyapunov exponent, and the fact that it is positive allows us to say, by definition, that the system (2.1) exhibits *chaos*. A system of dimension $>$ 1 will have a whole spectrum of characteristic Lyapunov exponents (LCE), and is chaotic if one of the LCE is greater than zero. (2) The (chaotic) evolution of (2.1) may be considered as random as coin tossing. Call x_n "heads" if it lies between 0 and 1/2, "tails" if it is between 1/2 and 1

Write the x_n's in base 2, so that $x_n = .d_n d_{n+1} d_{n+2} \ldots$, with each $d_i = 0$ or 1. Thus x_n is heads if $d_n = 0$, tails if $d_n = 1$. Now consider any possible sequence of heads and tails, say HHTHTTHTHH..., produced by coin flipping. It is easy to see that we can produce this same sequence with (2.1) by simply choosing x_0 appropriately. That is, any possible sequence of heads and tails corresponds to a particular choice of x_0, and we cannot distinguish the results of (random!) coin flipping from the evolution described by (2.1). *This is true in spite of the fact that (2.1) is perfectly deterministic.*

The thing that must be emphasized is that real physical systems exhibit such deterministic chaos. A system may be described by perfectly deterministic equations, and yet its dynamics may be as random as coin tossing. Such a system, because of its extreme sensitivity to initial conditions, is "indeterminable" in spite of its determinism. We can again use (2.1) to illustrate this point. If $x_0 = .d_1 d_2 d_3 \ldots d_n d_{n+1} \ldots$ then $x_n = .d_n d_{n+1} \ldots$ and, for large n, x_n obviously depends very sensitively on the precise value of x_0. In practice we cannot know x_0 precisely. And in any case any computer can deal only with a finite string of digits. As a practical matter, therefore, a chaotic system is ultimately *unpredictable* in detail.

In algorithmic complexity theory a sequence is said to be "random" if there exists no rule for producing the sequence simpler than just writing out the entire sequence. That is, there is no shorter program for generating the sequence than the trivial one of printing out the sequence bit by bit. The system (2.1) is known to be random in this sense for almost all x_0. For a chaotic system the best we can do, aside from appproximations, is simply to observe the system evolve; we cannot invent a simpler predictor, like a closed-form analytic solution. To paraphrase von Neumann, *the best description of a chaotic system is the system itself*.

At the other extreme are "regular" or non-chaotic systems. An obvious example is any periodic system. A more general type of regular motion is described by a *quasiperiodic system* in which any coordinate $x(t)$ of the system may be written as a discrete Fourier series

$$x(t) = \sum_{j}^{N} c_j \cos(\omega_j t + \theta_j) \tag{2.2}$$

Quasiperiodic systems are never chaotic. In other words, if a system is chaotic its Fourier spectra will have non-discrete, broadband components. Quasiperiodic systems are sometimes called "almost periodic" or "multiply-periodic."

An important feature of a quasiperiodic system is the *recurrence property*: for any t, and for any $\epsilon > 0$, there exists a T such that $|x(T) - x(t)| < \epsilon$. Therefore the value x(t) will be reached an infinite number of times. The frequency with which the function (2.2) has the value q is given by a theorem of Kac:[4,5] If the ω_j are linearly independent (i.e., incommensurate frequencies), then the mean frequency L(q) of the value q is

$$L(q) = (2\pi^2)^{-1} \iint_{-\infty}^{\infty} d\alpha\, d\eta\, \eta^{-2} \cos(q\alpha) \Big[\prod_{k=1}^{N} J_0(\alpha |c_k|) - \prod_{k=1}^{N} J_0\big(\sqrt{\alpha^2 + \eta^2 \omega_k^2}\, |c_k|\big) \Big] \tag{2.3}$$

If a quasiperiodic system has a spectrum composed of many incommensurate frequencies, the recurrence times may be extremely large and unobservable in practice. A simple example where recurrences are observable is provided by the Jaynes–Cummings model.[6] In this model a two-state atom interacts with a single mode of the electromagnetic field, and the rotating-wave approximation is made. If the atom is in the excited state at $t = 0$, and its transition frequency equals the field frequency, then the probability of its being excited at time t is

$$P(t) = \sum_{n=0}^{\infty} p_n \cos^2(\tfrac{1}{2}\Omega\sqrt{n+1}\,)t \tag{2.4}$$

where p_n is the probability that there are initially n photons in the field, and Ω is the Rabi frequency. The recurrence behavior of the (quasiperiodic) function (2.4) has been described in terms of "quantum collapse and revival,"[7] and experimental evidence for such behavior has been found.[8] Although it might be considered a "novel phenomenon,"[9] it can also be argued that there is nothing terribly surprising about it: *any quantum system with a purely discrete spectrum will display such "collapse and revival,"* at least if one waits long enough. We will return to quantum recurrences in Section 7, but for now let us move on to discuss chaotic versus quasiperiodic behavior in macroscopic, dissipative systems.

3. Chaotic Lasers

In 1978 Casperson[10] reported some peculiar behavior of a low-pressure He–Xe electric-discharge laser. For certain ranges of discharge current the output at 3.51 μm was oscillatory, even though the pumping and loss parameters were constant in time. For sufficiently large values of the current this "self-pulsing instability" resulted in apparently chaotic output, with a broadband spectrum. Casperson[11] performed numerical simulations of this behavior and noted that the self-pulsing instability could not be accounted for within a rate-equation analysis neglecting off-diagonal coherence in the Bloch equations for a two-state atom. Furthermore the pulsing was observed to be slow on the scale of the cavity transit time 2L/c, and so this *single-mode* instability is unrelated to mode-locked pulsing involving many longitudinal modes

Experiments by Abraham's group[12] revealed several "universal" routes to chaos associated with the Casperson instability, and numerical experiments on the Maxwell-Bloch equations showed qualitatively good agreement with the experiments.[13] These numerical studies confirmed that the chaotic behavior is explanable within the context of the *deterministic* Maxwell-Bloch equations for a single-mode, Doppler-broadened laser:[13]

$$\dot{u} = -(\Delta + \dot{\phi} - ks)v - \beta u \tag{3.1a}$$

$$\dot{v} = (\Delta + \dot{\phi} - ks)u - \beta v + \Omega(z_2 - z_1) \tag{3.1b}$$

$$\dot{z}_2 = R_2 - \gamma_2 z_2 - \tfrac{1}{2}\Omega v \tag{3.1c}$$

$$\dot{z}_1 = R_1 - \gamma_1 z_1 + \tfrac{1}{2}\Omega v \tag{3.1d}$$

$$\dot{\Omega} = -\gamma_c \Omega + K\int_{-\infty}^{\infty} ds W(s) v(s,t) \tag{3.1e}$$

$$\dot{\phi} = -(K/\Omega)\int_{-\infty}^{\infty} ds W(s) u(s,t) \tag{3.1f}$$

Here Δ is the frequency detuning of the field carrier frequency from the atomic transition frequency, and the off-diagonal decay rate β is 2π times the homogeneous linewidth (HWHM) of the transition. The electric field in the cavity is assumed to have the form $A(t)\cos[\omega t + \phi(t)]$, and $\Omega = \mu A/\hbar$, where μ is the transition dipole moment, is the Rabi frequency. γ_c is the field damping rate, determined mainly by the output coupling. z_2 and z_1 are the upper- and lower-level occupation probabilities, with corresponding pumping and decay rates R_j and γ_j, and u, v are the off-diagonal Bloch variables.[14] $ks = \omega s/c$ is the Doppler shift for an atom with velocity component s along the cavity axis, and W(s) is the (one-dimensional) Maxwell-Boltzmann velocity distribution function. The parameter $K = 2\pi N\mu^2\omega/\hbar$, where N is the number density of lasing atoms.

In order to accurately "resolve" the Maxwell-Boltzmann distribution, it is necessary to use ≈ 50 – 200 velocity groups, so that the system (3.1) on a computer is replaced by ≈ 10^2 ordinary differential equations. Figure 1 shows results for $\Delta = 0$, $R_1 = 0$, $R_2 = 8.5 \times 10^{-9}\beta$, $\gamma_c = 5.4\beta$, $\gamma_1 = 0.38\beta$, $\gamma_2 = .012\beta$, $\beta = 61$ MHz, $K = 6.4 \times 10^{23}$ sec^{-2}, and Doppler width $\delta\nu_D = 110$ MHz. (The numerical values of the parameters are discussed in Reference 13.) Figure 1a shows the computed intracavity intensity I(t) as a function of time, after the decay of initial transients. When R_2 is raised to $9.0 \times 10^{-9}\beta$, corresponding to a larger discharge current, the results are shown in Figure 1b. *Note that a period doubling has occurred.* Figures 1c and 1d show I(t) for $R_2 = 9.3$ and $9.4 \times 10^{-9}\beta$, respectively, revealing *more period doublings*. Slight further increases in R_2 produce more period doublings and eventually *chaos*. The results appear to be consistent, at least qualitatively, with the universality theory for the period doubling route to chaos.[1]

Figure 2 shows results for $R_2 = 5.6 \times 10^{-9}\beta$, $\Delta \neq 0$, and all the other parameters as in Figure 1. Power spectra of the field are shown for (a) $\Delta = 3.725\beta$, (b) $\Delta = 3.735\beta$, (c) $\Delta = 3.74\beta$, and (d) $\Delta = 3.75\beta$. In (a) there

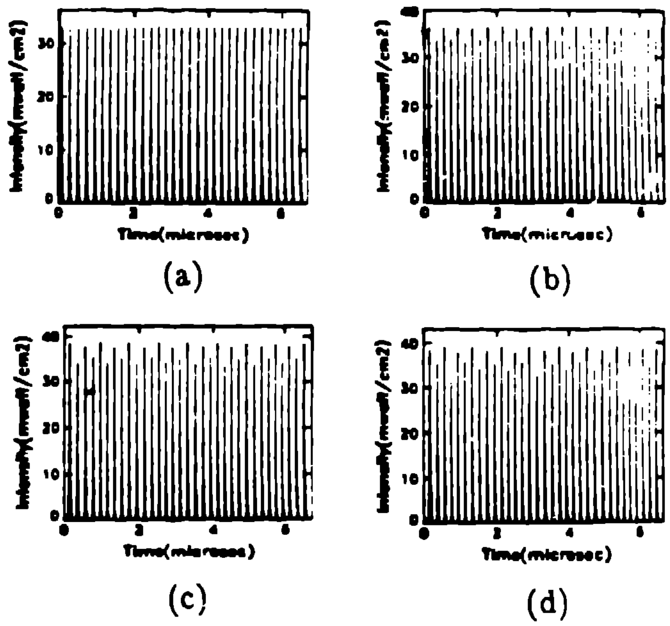


Figure 1. Period doubling to chaos as the pump rate is increased.

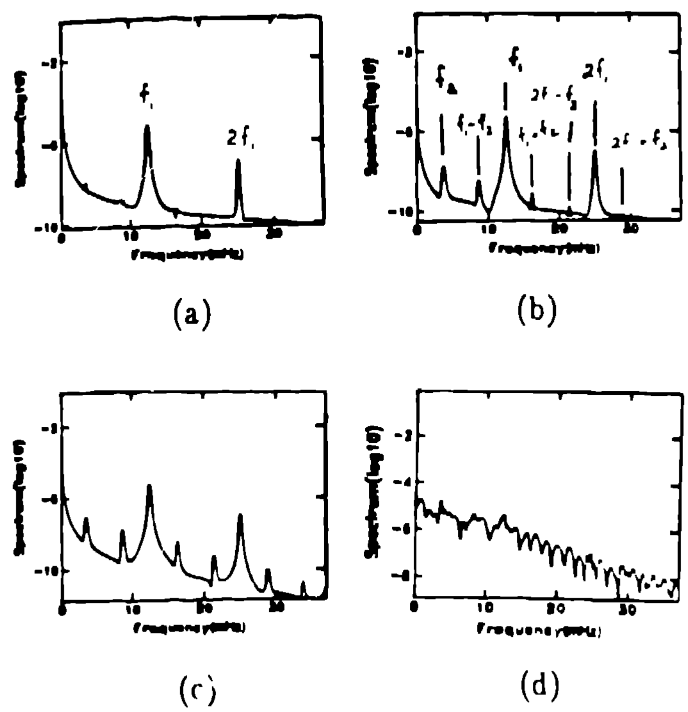


Figure 2 Two frequency route to chaos as the detuning is varied

is a single basic frequency and its harmonics in the field spectrum, but as the detuning Δ is increased there is the onset of two-frequency motion (b) followed by the broadband signature of chaos (d). Here we are observing the so-called two-frequency route to chaos, or the transition from quasiperiodic motion on a two-torus to chaos.[1]

We have also observed the route to chaos via "intermittency." In Figure 3, for instance, we show results for β = 38 MHz, $\gamma_1 = 1.8\beta$, $\gamma_2 = .057\beta$, $\gamma_C = 9\beta$, $K = 3.6 \times 10^{21}$ sec^{-2}, $\delta\nu_D$ = 110 MHz, $R_1 = 0$, and a variable pump rate R_2. Figures 3a – 3d are for R_2 = 1.2875, 1.29, 1.33, and 1.40 $\times 10^{-5}$ β, respectively. We have also observed "metastable chaos," i.e., long chaotic periods followed by abrupt transitions to quasiperiodic motion.

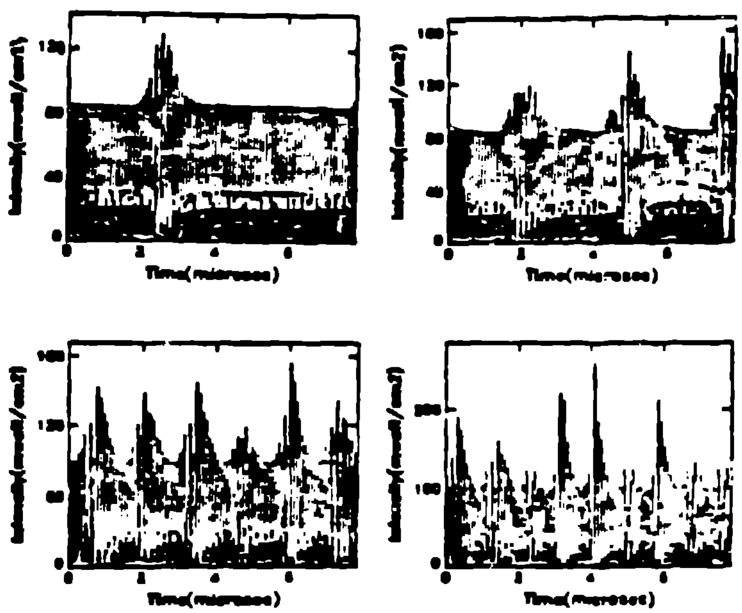


Figure 3. Development of chaos via intermittency.

These numerical studies may be summarized by saying that *we have observed the best-known routes to chaos for dissipative systems in the regime of the Casperson instability, and that the numerical and laboratory experiments appear to be consistent with each other.*

There have been many recent experimental and theoretical studies of chaos in laser devices with saturable absorbers, feedback, tilted mirrors, etc., and the interested reader will have no trouble finding papers to read on the subject. We have chosen to discuss briefly only the Casperson instability because it remains one of the few examples where the three prevalent routes to chaos in dissipative systems may be found in the same device.

4. Chaos in Optically Bistable Devices

A system is said to be bistable if it has two possible outputs for one and the same input. Optical bistability, which of course is of interest in connection with optical computers, generally refers to an optical system with two possible outputs for the same input intensity.[15,16] Consider, for instance, a laser beam injected into a cavity containing N absorbing two-state atoms per unit volume. For the case of homogeneous broadening, and exact resonance between the atoms and the field, we have the Maxwell-Bloch equations

$$\dot{v} = -\beta v + (\mu/\hbar)(A + A_0)z \tag{4.1a}$$

$$\dot{z} = -\gamma(z + 1) - (\mu/\hbar)(A + A_0)v \tag{4.1b}$$

$$\dot{A} = -\gamma_c A + (2\pi N\mu\omega)v \tag{4.1c}$$

where A_0 is the constant amplitude of the injected field and A is the amplitude of the intracavity field generated by the atoms. The steady-state solution of (4.1) gives the relation

$$X_0 = X + aX/(1 + X^2) \tag{4.2}$$

where $X = (A + A_0)/A_{SAT}$, $X_0 = A_0/A_{SAT}$, $A_{SAT} = (\hbar/\mu)\sqrt{\beta\gamma}$, and $a = c\alpha_0/2\gamma_c$, where $\alpha_0 = 4\pi N\mu^2\omega/\hbar\beta c$ is the line-center absorption coefficient. Figure 4 is a plot of X vs. X_0 for a = 25, showing that the total field in a cavity containing an absorbing medium can be a multivalued function of the injected field.[17,18]

Figure 5 shows a ring cavity configuration for an optically bistable

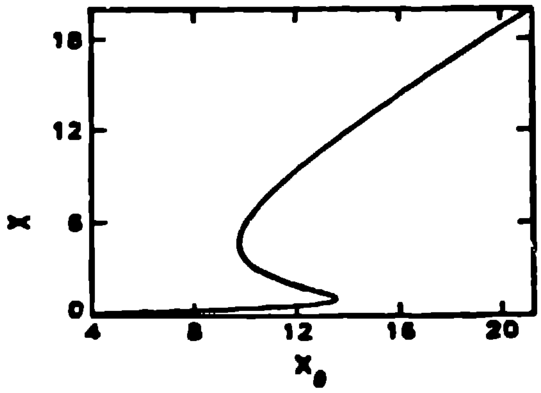


Figure 4. Plot of X vs. X_0 satisfying equation (4.2) for a = 25.

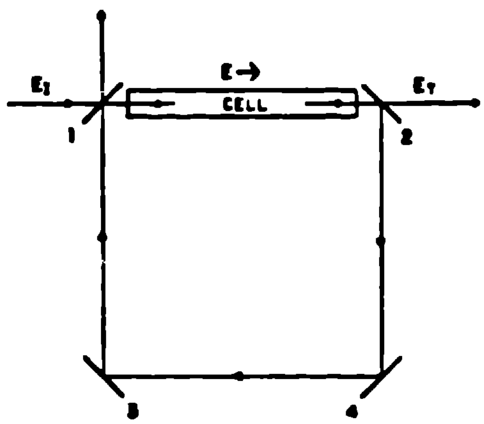


Figure 5. Ring cavity configuration for an optically bistable device.

device containing an absorbing cell. The possibility of chaos in such a system was first noted by Ikeda:[19] *within a range of input field intensities the transmitted field undergoes a period doubling route to chaos*. Experiments on both "all–optical" and "hybrid" devices have nicely supported this prediction.[20,21]

5. Chaotic Polarization of Light

One of the simplest problems of nonlinear optics, at least conceptually, is the coupling of two counter–propagating fields in a nonlinear medium. However, even such a "simple" system may have chaotic dynamics. Gaeta, *et al.*[22] predicted that the *polarization* of light in such a system may vary chaotically in time when the input intensity is sufficiently large. The equations considered by these authors are

$$\left[\frac{\partial}{\partial z} \pm \frac{1}{c}\frac{\partial}{\partial t}\right]E_i^{f,b} = \pm ik \sum_{j=x,y}\left[\chi_{ij}^{(0)}E_j^{f,b} + \chi_{ij}^{(\pm 2k)}E_j^{b,f}\right] \qquad (5.1)$$

and

$$\tau\frac{\partial \chi_{ij}}{\partial t} + \chi_{ij} = (A - B)(\vec{E}\cdot\vec{E}^*)\delta_{ij} + B(E_iE_j^* + E_i^*E_j) \qquad (5.2)$$

The superscripts f,b denote forward– and backward–propagating fields, respectively, and χ_{ij} is the electric susceptibility tensor. $\chi_{ij}^{(0)}$ and $\chi_{ij}^{(\pm 2k)}$ are the Fourier components of χ_{ij} at zero spatial frequency and spatial frequencies $\pm$ 2k, respectively. The Debye relaxation equation (5.2) applies to a Kerr medium characterized by the real constants A, B and a response time τ. For $\tau << L/c$, where L is the length of the Kerr medium, the medium response is effectively steady–state.

Numerical solutions of equations (5.1) and (5.2) show that when $B \neq 0$ the polarization of the transmitted light can be oscillatory when the total input intensity $(I_f + I_b)$ is large enough, and that for sufficiently large input intensities the polarization can vary chaotically in time, as shown in Figure 6. Such polarization chaos can occur while the total transmitted intensity is constant in time.[22]

Polarization chaos is not unique to Kerr media, but may occur whenever there is a nonlinear interaction of vector fields. Gauthier, *et al.*[23] have observed polarization instabilities and chaos in an experiment employing counter-propagating dye laser beams in sodium vapor.

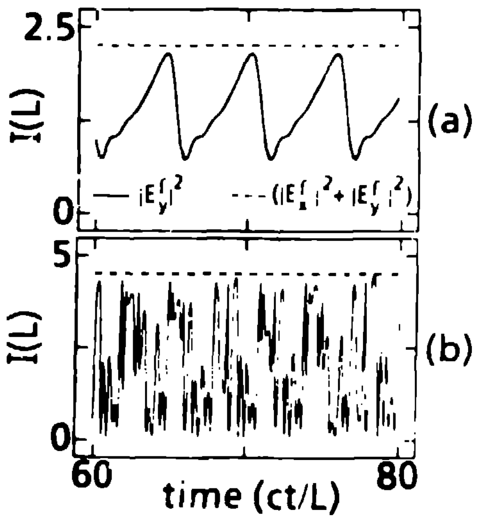


Figure 6. Time evolution of the total transmitted intensity and the intensity of one component of polarization for $\tau << L/c$ and unequal input intensities. (See Reference 22.)

6. <u>Chaos in the Jaynes–Cummings Model</u>

The Schrödinger equation for a two-state atom of transition frequency ω_0 in an electric field E may be written in the well-known Bloch form[14]

$$\dot{x} = -\omega_0 y \tag{6.1a}$$

$$\dot{y} = \omega_0 x + (2\mu/\hbar)Ez \tag{6.1b}$$

$$\dot{z} = -(2\mu/\hbar)Ey \tag{6.1c}$$

If N two-state atoms per unit volume are contained in a cavity supporting a single field mode, and if we assume the atoms are all lumped together within a wavelength, then we can write the Maxwell equation

$$\ddot{E} + \omega_0^2 E = 4\pi N\mu x \tag{6.2}$$

We are assuming for simplicity that the cavity mode is exactly resonant with the atoms.

It is convenient to define the dimensionless parameter $\beta = 8\pi N\mu^2/\hbar\omega_0$. For $\mu = 1$ D and $\omega_0 = 10^{15}$ sec^{-1}, we have $\beta = 2.4 \times 10^{-23}$N. It is found that the system (6.1) plus (6.2) exhibits chaos when $\beta \approx 1$ or larger.

The system under consideration is just an extension of the Jaynes–Cummings model mentioned earlier to the case of N atoms per unit volume, *except that the rotating-wave approximation (RWA) is not made*. And in the present model the field is treated classically, i.e., we are dealing with a semiclassical approximation to the usual Jaynes–Cummings model.

This example of chaos in an AMO system is very interesting, for it is a truly fundamental model. (Note that we have not included any damping terms of any kind.) It is also interesting because, when *the ubiquitous RWA is made, the dynamics are predicted to be quasiperiodic*. We reported these results in 1983,[24] at a time when we were first learning about chaos and thinking about how it might manifest itself in fundamental models of light–matter interactions. However, it was soon pointed out to us that the same model had been considered at least seven years earlier by Belobrov, Zaslavskii, and Tartakovskii![25] These authors used a slightly different form of the interaction Hamiltonian, but for the present discussion this difference is inconsequential.[26]

The present example can also be thought of in terms of a single two-state atom interacting with a single cavity mode; chaos results if the atom–field coupling strength is large enough. But then we can no longer trust the semiclassical approximation, and must address questions of "quantum chaos."

7 Quantum Chaos

The subject of "quantum chaos" is concerned with how, if at all, classical chaos carries over into quantum theory. It has been a controversial subject, and much has been written about it, and I cannot in the space allotted review the field in any serious way. Instead I will present some thoughts based mainly on the consideration of a few model systems.

One area of investigation concerns properties of eigenvalues. Energy level distributions can exhibit sensitivity to a nonlinear parameter when the corresponding classical system goes chaotic.[27] The energy levels can have complicated level crossings, and their spacings can follow a Wigner distribution,[28] but it is not obvious precisely how these measures of "quantum chaos" correspond to the classical chaos in the sense of exponential sensitivity on initial conditions and the consequent loss of predictability.

The most obvious question in the latter context is whether the wave function can evolve chaotically in time, with the kind of exponential sensitivity to initial conditions exhibited by classical chaotic trajectories. For systems with purely discrete energy spectra the answer is clearly that it cannot. For such systems we can write

$$\psi(t) = \sum_n |c_n| e^{-i(E_n t + \theta_n)} \phi_n \tag{7.1}$$

where the E_n are the energy eigenvalues and the ϕ_n are the corresponding eigenvectors. Then

$$||\psi(t_2) - \psi(t_1)|| = \sum_n |c_n|^2 [1 - \cos E_n(t_2 - t_1)] \tag{7.2}$$

The quasiperiodicity of this norm means that two state vectors cannot separate exponentially with time, i.e., $\psi(t)$ cannot evolve chaotically.

The fact that $\psi(t)$ is quasiperiodic implies the *quantum recurrence theorem:*[29] if $\psi(t_0)$ is the state vector at time t_0, and ϵ is any positive number, there exists a time T such that $||\psi(T) - \psi(t_0)|| < \epsilon$. This quantum recurrence theorem is the analogue of the classical Poincare' recurrence theorem, which says that any initial point in the phase space of a system of finite volume is recurrent.

This analogy immediately raises a simple but often overlooked point: the fact that the state vector is recurrent should not by itself be used as an argument against quantum chaos, just as the Poincare' recurrence theorem cannot be used to argue that there is no classical chaos! For the mere *recurrence* of some initial state does not imply regular behavior (predictability), any more than the recurrence of "heads" in coin tossing means there is no randomness there.

The recurrence *per se*, then, cannot be used as proof that quantum chaos is impossible, as sometimes argued.[30] These proofs begin with the *assumption* of quasiperiodicity to conclude that the dynamics are recurrent, but *it is the assumption of quasiperiodicity itself that renders quantum chaos impossible in the sense of a positive Lyapunov exponent.* For we have already noted that quasiperiodic means regular and predictable, never chaotic. Thus I agree with the conclusions of the argument against quantum chaos in systems with discrete energy (or quasienergy) spectra, if not with the argument itself.

(Having said that, it must be noted that quantum recurrence and classical Poincare' recurrence are rather different things. Whereas nearby points in classical phase space may have quite different recurrence times, there can be many similar quantum states with similar recurrence times[31].)

An argument sometimes heard against the possibility of quantum chaos is that the Schrödinger equation is linear and therefore cannot allow chaos, which occurs only in nonlinear (or piecewise linear) systems. This argument is specious because, although the time-dependent Schrödinger equation can be written as a set of linear ordinary differential equations for probability amplitudes, this set is generally *infinite*, and such infinite sets of linear equations can admit chaos. Indeed it is possible to transform a finite-dimensional *nonlinear* system into an infinite-dimensional *linear* system.

Another argument against the possibility of chaotic time evolution of the state vector is that the scalar product $<\psi_1(t)|\psi_2(t)>$ of two state vectors is invariant in time, and therefore if two states are similar at t = 0, they will remain similar.

At this point we must acknowledge the important conceptual differences between quantum mechanical state vectors and classical trajectories. In particular, the state vector is not the quantum analogue of a classical trajectory but rather corresponds, roughly speaking, to a whole *ensemble* of such trajectories. So the thing to compare with the time evolution of the state vector is not the path in phase space of some trajectory, but rather the evolution of some initial distribution in classical phase space. The question then arises: *can a classical distribution in phase space evolve chaotically in time in the sense of exponential sensitivity to the initial distribution?*

Consider the Liouville equation for the phase space distribution ρ: $\partial\rho/\partial t = \{H,\rho\}$. From this equation it is easy to show, for continuous distributions, that the overlap of any two distributions is constant in time.

$$\frac{\partial}{\partial t}\int d^N q\, d^N p\, \rho_1 \rho_2 = 0 \tag{7.3}$$

This is analogous to $(\partial/\partial t)\langle\psi_1|\psi_2\rangle = 0$, and suggests that classical *distributions* of trajectories may have less sensitivity to initial conditions than the trajectories themselves.

Of course the more appropriate distribution function for our discussion is the Wigner distribution, which can be regarded as the quantum analogue of the classical ρ. For a few models which have been studied,[32,33] the Wigner distribution appears to vary more regularly in time than the classical distribution.

In the path-integral formulation all the classical paths contribute to the time evolution of a quantum system. Heuristically, we can expect the summation over the irregular classical trajectories to smooth out the classically chaotic behavior.

For systems driven by time-periodic forces another aspect of the quantum suppression of classical chaos emerges. In the classical description the meandering of chaotic trajectories in phase space leads to an effectively diffusive behavior in which the energy, averaged over trajectories, grows approximately linearly with time. In the quantum description of a few model systems, however, it is found that the dynamics are restricted over a relatively small number of eigenstates, and the energy does not grow linearly with time for long times, but tends instead to saturate. For the model of the kicked pendulum this has been explained by analogy to Anderson localization.[34] The basic idea is to transform the dynamics to the form

$$(W_0 + T_n)c_n = \sum_{r \neq 0} W_r c_{n+r} \tag{7.4}$$

Here c_n is the amplitude for the nth eigenstate and $T_n = \tan[\frac{1}{2}(\omega T - n^2\tau)]$, where ω belongs to the quasienergy spectrum of the periodically driven system with driving period T, $\tau = \hbar T/m\ell^2$, where m and ℓ are the mass and length of the pendulum; the coefficients W_r need not concern us here.

Equation (7.4) is of the same form as the Schrodinger equation in the tight binding model of Anderson localization. The T_n in that case are site energies, which are taken to be independent random variables. This assumed randomness of the site energies is responsible for the Anderson localization, i.e., the absence of quantum diffusion over the lattice. In (7.4), in contrast, the T_n are not assumed random variables, but are determined by the driving period and the energy levels (proportional to n^2) of the unperturbed pendulum.

But the sequence $\{T_n\}$ *does* exhibit randomness, much like the sequence (2.1). This is connected with the n^2 in the definition of T_n, which in turn is connected to the energy level structure of the pendulum. This random property, resulting from the quantum energy level structure of the unperturbed system, provides the analogy to Anderson localization, and an explanation for the absence of diffusive energy growth in the quantum description of the periodically kicked pendulum.

This analogy to Anderson localization is very elegant, but it appears to apply only to unbounded quantum systems, as in the kicked pendulum where E_n increases as n^2.

A number of classical analyses of atomic and molecular systems in applied fields have shown that chaos is an important mode of behavior in the classical description of such processes. For instance, we have found that the appearance of chaos in models of infrared multiple-photon excitation (MPE) of large molecules can be responsible for a diffusive energy growth and the fact that the MPE process is found experimentally to be fluence-dependent.[35-37]

The most extensively studied system in this connection, of course, is the microwave ionization of highly excited hydrogen atoms.[38] For this system the classical theory provides remarkably good predictions for ionization probabilities, except for certain microwave frequencies. Not surprisingly, quantum theory provides even more accurate predictions. The big difference is that there is chaos in the classical theory but not in the quantum.

The lack of any evidence for quantum chaos, i.e., for anything but regular evolution of the wave function, seems to be regarded as some sort of mystery. In fact one prominent researcher has expressed the view that, since chaos is an observed fact of Nature, and quantum theory apparently does not admit chaos, we must be faced with a failure of quantum theory, and that we are "headed for a revolution." In the remainder of this lecture I would like to argue that no revolution will be necessary.

For a system like the kicked pendulum we should not be terribly surprised that the classical and quantum dynamics are, for sufficiently long times, quite different: as we move up the energy scale the energy level spacings increase monotonically, and the higher we go, the more the distinctly quantum features will manifest themselves. Note that the monotonic growth of energy with quantum number n is a crucial point in the analogy to Anderson localization.

The situation is different, of course, for an atom or molecule, where the energy spectrum is discrete only up to an ionization or dissociation limit. Detailed comparisons between classical and quantum dynamics have been made only for a few such systems, but such comparisons have shown, by and large, that the two theories are in good agreement in their predictions of ionization or dissociation probabilities.[38–40] Here again the situation does not necessitate any radical departure from conventional theory, as I will illustrate with results obtained for the driven Morse oscillator.[39–41]

The Hamiltonian for the driven Morse oscillator is

$$H = p^2/2m + D(1 - e^{-\alpha x})^2 - dxE_L\cos(\omega_L t) \qquad (7.5)$$

D and α are the dissociation energy and range parameter, respectively, of the Morse potential, and d is the dipole moment gradient. The classical equations of motion may be written in the scaled form

$$d^2X/d\tau^2 = (4/B^2)(e^{-X} - e^{-2X}) + 2K\cos(\mu\tau) \qquad (7.6)$$

where $\tau = (DB^2/\hbar)t$, $X = \alpha x$, $\mu = \hbar\omega/DB^2$, $K = dE_L/\alpha DB^2$, and the dimensionless parameter $B = \hbar\alpha/\sqrt{2mD}$. We have mainly used parameters corresponding to the HF molecule, for which there are 24 bound states of the Morse potential. In terms of the same scaled variables the Schrodinger equation is

$$i\partial\psi/\partial\tau = -\partial^2\psi/\partial X^2 + B^{-2}(1 - e^{-X})^2\psi - KX\cos(\mu\tau)\psi \qquad (7.7)$$

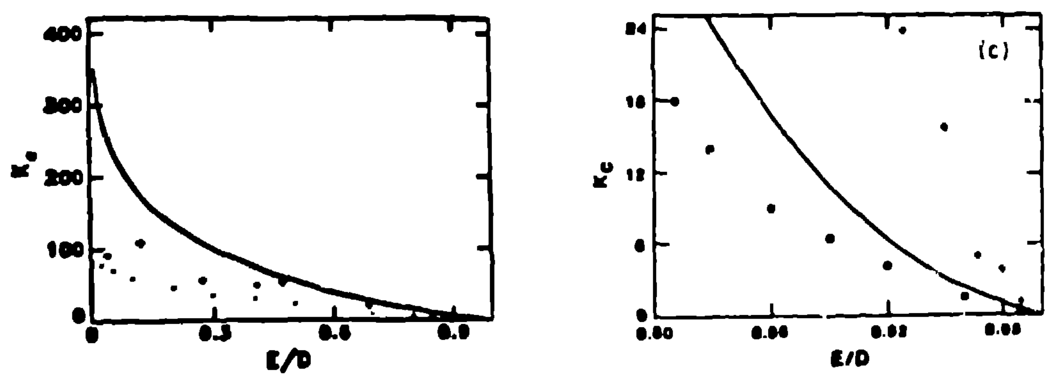
To compare the classical and quantum predictions, we do the following. In the quantum theory we solve (7.7) numerically, and compute the probability

$$P_D(t) = \sum_n |\langle\psi_n|\psi(t)\rangle|^2 \tag{7.8}$$

summed over the *discrete* eigenfunctions $|\psi_n\rangle$. The dissociation probability at time t is then $1 - P_D(t)$. In the classical theory we solve (7.6) for an ensemble of classical trajectories, and define the dissociation probability as the *fraction* of trajectories that escape the Morse well.[39,40]

In driven Hamiltonian systems, Chirikov's classical resonance overlap criterion[42] may be used to predict the amplitude of the driving force necessary for the onset of global chaos. In systems with dissociation or ionization, however, the term "chaos" is somewhat ambiguous, for the computation of Lyapunov exponents requires, in principle, a $t \to \infty$ limit. In dissociating (or ionizing) systems, however, the dissociation may occur very quickly, within a few cycles of the driving force, and it is not clear how to rigorously define the "transient chaos" in the pre-dissociation dynamics. For such systems we prefer therefore to phrase the question of "quantum chaos" as follows: *How, if at all, does classical resonance overlap manifest itself quantum mechanically?*

First let us note that the simplest form of the resonance overlap criterion allows us to predict fairly well the critical field strength K_c necessary for dissociation. Figure 7 compares K_c predicted by the classical resonance overlap criterion ( ), classical dynamics (·), and quantum theory (+) as a function of the initial energy E of the unperturbed Morse oscillator. Figure 7a is for the case of an N = 1 classical nonlinear resonance, whereas Figure 7b is for an N = 4 resonance. For the Morse oscillator such resonances occur when the laser frequency $\omega_L = N\omega_0\sqrt{1 - E/D}$, where $\omega_0 = \sqrt{2D\alpha^2/m}$ is the natural oscillation frequency for the nearly harmonic motion near the bottom of the well. Note that the three predictors for K_c plotted in Figure 7 come into better agreement as E/D increases, as might be expected. We have also found that the differences between the classical and quantum predictions are most pronounced near higher order classical resonances (N > 1) and quantum multiphoton resonances.[41] Details and explanations may be found in References 39 and 40.



(a) (b)

Figure 7. Predictions of the critical field strength necesary for dissociation for laser frequencies at (a) N = 1 and (b) N = 4 nonlinear resonances.

The width of a classical resonance turns out to be proportional to the square root of the applied field amplitude. Based on the simplest semiclassical arguments we conclude that the number of quantum levels coupled by the field should also be proportional to the square root of the field amplitude, and this surmise is corroborated pretty well by our numerical experiments.[39,40]

Since the width of a classical resonance corresponds to a spread Δn of the number of quantum levels mixed by the field, an overlapping of classical resonances is associated with the mixing of a large number of quantum levels. When this happens *the quantum dynamics is complicated and can mimic the "chaotic" classical dynamics, but the quantum dynamics are quasiperiodic, not chaotic.*[39,40] Classically, resonance overlap results in diffusive motion in phase space, leading to dissociation. Quantum mechanically, it is the spread of population with increasing field strength that gives rise to dissociation.

We have argued for several years that quantum systems with a relatively small number of incommensurate energy levels or frequencies can mimic classical chaos in their "randomness" and in the time it takes to observe recurrence phenomena.[39,40,43] In particular, the wave function can exhibit properties that are consequences of chaos – like broadband spectra, "decaying" correlations, and certain ergodic properties[43] – without evolving chaotically in time.

Of course the view that quantum chaos is generally not possible, and that quantum dynamics can only *mimic* chaotic behavior in some circumstances, will have to be modified if just one example of true quantum chaos is discovered. Fox and Eidson[45] have argued that the chaos in the Jaynes–Cummings model of Section 6 is an example of *quantum* chaos. In my opinion this is not the case because the field has not been quantized. Alekseev and Berman[46] apparently have found an example of quantum chaos in a system with stationary coherent states, where there is the maximum possible correspondence at all times with the classical limit. If this is true quantum chaos, it must nevertheless be acknowledged that such systems are highly atypical.[46]

I would like to thank Dr. P.I. Belobrov and the organizing committee for providing me with the opportunity to meet them and some of the other Soviet researchers whose work I greatly admire.

REFERENCES

1. For a review see, for instance, P.W. Milonni, M.-L. Shih, and J.R. Ackerhalt, *Chaos in Laser Matter Interactions* (World Scientific, Singapore, 1987).

2. E.R. Buley and F.W. Cummings, Phys. Rev. A1454, 134 (1964).

3. See, for instance, A. Wolf, J.B. Swift, H.L. Swinney, and J.A. Vastano, Physica 16D, 285 (1985).

4. M. Kac, Am. J. Math. 65, 609 (1943).

5. P. Mazur and E. Montroll, J. Math. Phys. 1, 70 (1960).

6. E.T. Jaynes and F.W. Cummings, Proc. IEEE 51, 89 (1963).

7. J.H. Eberly, N.B. Narozhny, and J.J. Sanchez Mondragon, Phys. Rev Lett. 44, 1323 (1980).

8. G. Rempe and H. Walther, Phys Rev Lett 58, 353 (1987).

9. S Haroche and D Kleppner, Physics Today, January, 1989, p 24

10. L.W. Casperson, IEEE J. Quantum Electron. QE-14, 756 (1978).

11. L.W. Casperson, in *Laser Physics*, ed. by J.D. Harvey and D.F. Walls, Lecture Notes in Physics 182 (Springer-Verlag, Berlin, 1983).

12. N.B. Abraham, et al., ibid.

13. M.-L. Shih, P.W. Milonni, and J.R. Ackerhalt, J. Opt. Soc. Am. B2, 130 (1985); Opt. Commun. 49, 155 (1984).

14. L. Allen and J.H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, N.Y., 1975).

15. H.M. Gibbs, *Optical Bistability: Controlling Light with Light* (Academic Press, N.Y., 1985).

16. E. Abraham and S.D. Smith, Rep. Prog. Phys. 45, 815 (1982).

17. A. Szöke, V. Daneau, J. Goldhar, and N.A. Kurnit, Appl. Phys. Lett. 15, 376 (1969).

18. S.L. McCall, Phys. Rev. A9, 1515 (1974).

19. K. Ikeda, Opt. Commun. 30, 257 (1979).

20. H. Nakatsuka, S. Asaka, H. Itoh, K. Ikeda, and M. Matsuoka, Phys. Rev. Lett. 50, 109 (1983).

21. H.M. Gibbs, S.L. McCall, and T.N.C. Venkatesan, Phys. Rev. Lett. 36, 1135 (1976).

22. A.L. Gaeta, R.W. Boyd, J.R. Ackerhalt, and P.W. Milonni, Phys. Rev. Lett. 58, 2432 (1987).

23. D.J. Gauthier, M.S. Malcuit, and R.W. Boyd, Phys. Rev. Lett. 61, 1827 (1988).

24. P.W. Milonni, J.R. Ackerhalt, and H.W. Galbraith, Phys. Rev. Lett. 50, 966 (1983).

25. P.I. Belobrov, G.M. Zaslavskii, and G. Th. Tartakovskii, Sov. Phys. JETP 44, 945 (1976).

26. J.R. Ackerhalt and P.W. Milonni, J. Opt. Soc. Am. B1, 116 (1984).

27. I.C. Percival, J. Phys. B6, 1229 (1973).

28. M.V. Berry, Phil. Trans. Roy. Soc. London A287, 237 (1977).

29. P. Bocchieri and A. Loinger, Phys. Rev. 107, 337 (1957).

30. T. Hogg and B.A. Huberman, Phys. Rev. Lett. 48, 711 (1982).

31. A. Hobson, *Concepts in Statistical Mechanics* (Gordon and Breach, N.Y., 1971).

32. H.J. Korsch and M.V. Berry, Physica 3D, 627 (1981).

33. J.S. Hutchinson and R.E. Wyatt, Phys. Rev. A23, 1567 (1981).

34. D.R. Grempel, R.E. Prange, and S. Fishman, Phys. Rev. A29, 1639

(1984).

35. J.R. Ackerhalt, H.W. Galbraith, and P.W. Milonni, Phys. Rev. Lett. 51, 1259 (1983).

36. J.R. Ackerhalt and P.W. Milonni, Phys. Rev. A34, 1211 (1986).

37. J.R. Ackerhalt and P.W. Milonni, Phys. Rev. A37, 1552 (1988).

38. See, for instance, G. Casati, I. Guarneri, and D.L. Shepelyansky, IEEE J. Quantum Electron. QE-24, 1420 (1988), and references therein to experiments of J.E. Bayfield and P.M. Koch.

39. M.E. Goggin and P.W. Milonni, Phys. Rev. A37, 796 (1988).

40. M.E. Goggin and P.W. Milonni, Phys. Rev. A38, 5174 (1988).

41. R.B. Walker and R.K. Preston, J. Chem. Phys. 67, 2017 (1977).

42. B.V. Chirikov, Phys. Rep. 52, 263 (1979).

43. P.W. Milonni, J.R. Ackerhalt, and M.E. Goggin, Phys. Rev. A35, 1714 (1987).

44. D.L. Shepelyansky, Physica 8D, 208 (1983).

45. R.F. Fox and J. Eidson, Phys. Rev. A34, 482 (1986); A34, 3288 (1986).

46. K.N. Alekseev and G.P. Berman, Sov. Phys. JETP 61, 569 (1985).