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## Experimental Evidence of Subharmonic Bifurcations, Multistability, and Turbulence in a $Q$-Switched Gas Laser


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Subharmonic bifurcations, generalized multistability, and chaotic behavior were found experimentally in a $Q$-switched $CO_2$ laser operating at 10.6 $\mu$m. Jumps between two strange attractors lead to a low-frequency ($1/f$ type) divergence in the power spectrum. This is the first experimental evidence of these phenomena in a quantum-optical molecular system. A theoretical model is also presented whose results are in good agreement with the experimental data.



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Over the past years, the laser has been a test bench for many conjectures in nonequilibrium statistical mechanics. Measurements of photon statistics around laser threshold suggested a set of analogies between stationary quantum optical devices and thermodynamical phase transitions.[1] Furthermore, evidence of transient anomalous fluctuations[2] introduced the new concept of nonstationary statistics, later extended to other physical systems.[1,3,4] Recently, interest has arisen in higher-order bifurcations of quantum optical dynamical systems, which may eventually lead to chaos. Theoretical models have been formulated,[5] and an experiment has been performed on a hybrid electro-optical device.[6]

Here we report the first experimental evidence of multiple subharmonic bifurcations, eventually leading to chaos, in a quantum optical molecular system.[7] Specifically, we show these effects in a $Q$-switched $CO_2$ laser operating at the 10.6 $\mu$m $P(20)$ line. Furthermore we give evidence of the coexistence of independent basins of attraction in the phase space (generalized multistability) which may lead in suitable situations to the appearance of low-frequency divergences in the power spectrum ($1/f$ noise).[8] We have chosen a $CO_2$ laser system because the relaxation time $1/\gamma_\parallel$ of the population inversion is much larger ($1/\gamma_\parallel = 0.4$ ms) than the memory time $1/\gamma_\perp$ of the induced dipoles[9] ($1/\gamma_\perp = 10^{-8}$ s), thus reducing the single-mode dynamics to the coupling between two degrees of freedom, namely photon population $n$ and molecular population inversion $\Delta$. Introducing within the cavity a time-dependent perturbation by an electro-optical modulator driven by a sinusoidal frequency, we have a nonautonomous differential system in $n$ and $\Delta$ amounting to the crucial three degrees of freedom that have been shown to be the necessary condition for the onset of chaos.[10] As shown later, the relevant range of modulation frequencies is correlated to the $\gamma_\parallel$ value; hence the choice of $CO_2$ laser has put the working frequency range in the easily accessible 50–130-kHz region.

The coupled field-molecules equations for a single-mode laser lead, after adiabatic elimination of the polarization, to the following rate equations:

$$\dot{\Delta} = R - Gn\Delta - \gamma_\parallel \Delta, \quad \dot{n} = 2Gn\Delta - K(t)n, \tag{1}$$

where

$$G = \omega\mu^2/\hbar\epsilon_0\gamma_\perp V = 0.25\times10^{-4}\ \mathrm{s}^{-1} \qquad (2)$$

is the coupling constant (SI units) including the frequency $\omega$ and the dipole matrix element $\mu$ of the $P(20)$ transition and the collisional broadening rate $\gamma_\perp$, $R$ is the pump rate, $V$ the cavity volume, and

$$K(t) = K_1(1 + m\cos\Omega t) \qquad (3)$$

is the cavity damping rate, modulated by the inserted electro-optical device. A linear perturbation analysis around the steady-state values

$$\bar{\Delta} = K_1/2G, \quad \bar{n} = 2R/K_1 - \gamma_\parallel/G, \qquad (4)$$

yields a linearized frequency value $\Omega_0/2\pi \simeq 43$ kHz for the following typical parameters: spontaneous emission rate $\gamma_{sp} = 1.3\times10^3\ \mathrm{s}^{-1}$, and $K_1 = 3\times10^7\ \mathrm{s}^{-1}$ (corresponding to a cavity length of 2 m with losses of 20% per pass), and for a pumping rate $R$ such that $\bar{n} \simeq \gamma_\parallel/G \simeq 10^8$ (corresponding to a dc power output of 50 $\mu$W). We select for the two modulation parameters $m$ and $\Omega$

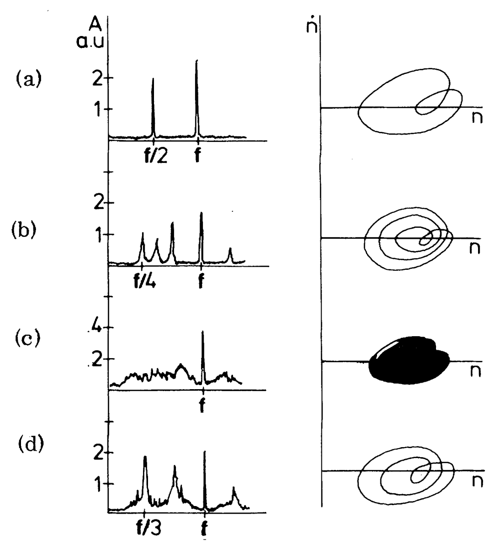


FIG. 1. Experimental phase-space portraits ($\dot{n}-n$) (right side) and the corresponding frequency spectra (left side) for different modulation frequencies $f$. (a) $f$ = 62.75 kHz. Period-two limit cycle and corresponding $f/2$ subharmonic. (b) $f$ = 63.80 kHz. Period-four limit cycle and $f/4$ subharmonic. (c) $f$ = 64.00 kHz. The phase-space portrait shows a strange attractor (the oscilloscope spot could not resolve single windings). The power spectrum is a quasicontinuous one with a small peak at the modulation frequency (see the scale change with respect to previous figures). (d) $f$ = 64.13 kHz. Period-three limit cycle and $f/3$ subharmonic.

ranges which have been shown to be relevant in previous studies of driven nonlinear oscillators.[8,11] Specifically, we set $K_1$ consistently below the maximum damping rate $K_0 = GR/\gamma_\parallel$ compatible with the fixed pump rate $R$. It is easily shown that the linearized eigenfrequency is

$$\Omega_0 \simeq (\gamma_\parallel K_1)^{1/2}, \qquad (5)$$

provided we choose $K_1 \approx K_0/2$ and a modulation depth $m$ sufficiently small to have

$$(1/K_1)(dK/dt) = \Omega m < \gamma_\parallel. \qquad (6)$$

The driving frequency $f = \Omega/2\pi$ is chosen to vary in the region from $\Omega_0/2\pi$ on, that is, from 40 to 150 kHz. As a consequence, $m < \gamma_\parallel/\Omega \simeq 10^{-2}$. We have explored modulation values between 1% and 5%. A complete state diagram would yield the dynamical features for all possible values of the modulation parameters $m$ and $\Omega$. However, the strip $m = 1\%-5\%$ does not display $m$ dependence; therefore we limit ourselves to giving experimental results at $m = 1\%$ for various $\Omega$ values.

The experimental setup consists of a $CO_2$ laser carefully stabilized against thermal and acoustic disturbances, with the discharge current stabilized better than $1/10^3$. No long-term stabilization was necessary. The electro-optical modulator was a CdTe, antireflex-coated, 6-cm-long crystal, with an absorption less than 0.2%. The laser cavity includes also a $\lambda/4$ plate and a beam expander, both coated to limit the total losses per pass to 20%. The laser output is detected on a fast (2.5-ns rise time) pyroelectric detector whose current, proportional to the photon number $n(t)$, is sent together with its time derivative $\dot{n}(t)$ to an $x$-$y$ scope, in order to have the phase-space portrait $(n, \dot{n})$. The detector is also sent to a Rockland spectrum analyzer to measure the

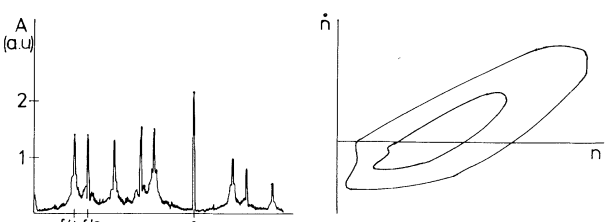


FIG. 2. $f$ = 63.85 kHz. Experimental evidence of generalized multistability (coexistence of two independent attractors). The power spectrum shows that those attractors correspond to $f/3$ and $f/4$ subharmonic bifurcations, respectively; in phase space, the multipole windings merged within the thickness of the phase portrait contour.

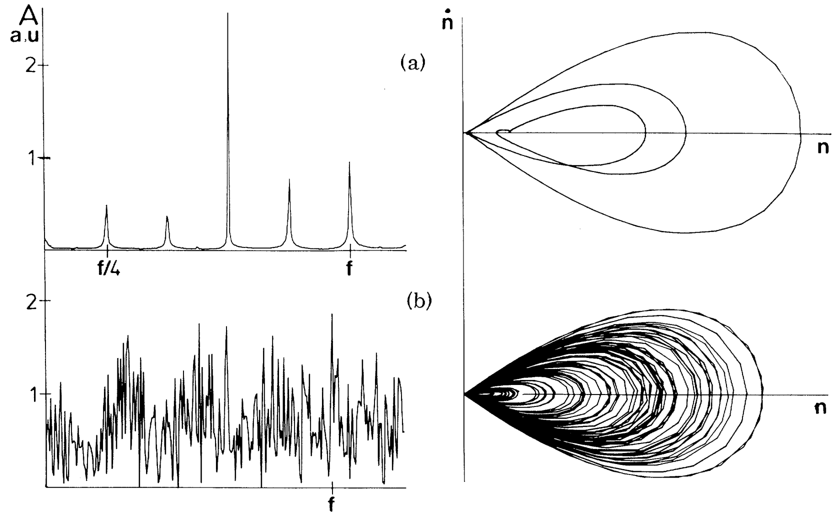


FIG. 3. Computer plots for the parameter values $\gamma_{\parallel} = 10^3$ s$^{-1}$, $K_1 = 7\times 10^7$ s$^{-1}$, $m = 2.0\times 10^{-2}$, $GR = 2.0\times 10^{11}$. (a) $f$ = 64.33 kHz. Subharmonic bifurcation $f/4$, as in the experiment of Fig. 1(b). (b) $f$ = 78.8 kHz, $m = 3\times 10^{-2}$. Strange attractor and broad spectrum corresponding to a chaotic solution.

power spectra. The limited range (up to 100 kHz) of the spectrum analyzer has limited the frequency range explored thus far. We show later that interesting bifurcations are also expected in the 130-kHz domain for which we do not have experimental data.

In Fig. 1 we show experimental data in a narrow region between 62.7 and 64.25 kHz where various bifurcations occur. This region is limited above and below by wide intervals with stable single-period limit cycles. Fig. 1(a) shows the $f/2$ bifurcation at $f = 62.7$ kHz, Fig. 1(b) the $f/4$ case for $f = 63.8$ kHz; Fig. 1(c) shows the strange attractor and a broad-band spectrum for $f = 64.0$ kHz; and Fig. 1(d) shows the $f/3$ case for $f = 64.2$ kHz. As one can see, in this interval there is no Feigenbaum sequence.[12] Furthermore at $f = 63.85$ kHz a new feature appears, namely the coexistence of two independent stable attractors, one of period 4 ($f/4$) and the other of period 3 ($f/3$) (Fig. 2). Notice that the driving frequency is intermediate between those of Figs. 1(b) and 1(d). Therefore the chaotic behavior of Fig. 1(c) can be considered as a merging of those two attractors. This bistable situation has nothing to do with the common optical bistability[13] where two dc output amplitude values appear for a single dc driving amplitude. We call this coexistence of two attractors "generalized bistability."

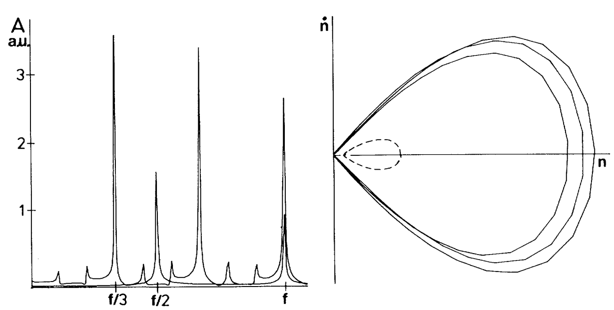


FIG. 4. Theoretical generalized bistability; $f$ = 119.0 kHz, $m = 2.0\times 10^{-2}$. The phase-space portrait shows the existence of two independent attractors, corresponding to the subharmonic frequencies $f/2$ (dashed line) and $f/3$ (continuous line); relative spectra are superimposed. It must be noted that one attractor remains inside the other as in the experiment of Fig. 2. If initial conditions are properly changed, a third attractor is found with a subharmonic frequency $f/10$ (not plotted for the sake of simplicity). Initial conditions: $n_0 = 4\times 10^8$, $\dot{n}_0 = 0$ (dashed), $n_0 = 2\times 10^5$, $\dot{n}_0 = -2\times 10^6$ (continuous).

In Figs. 3 and 4 we report the theoretical equi-

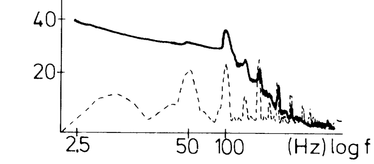


FIG. 5. Experimental power spectra in the case of two attractors, stable (dashed line), and strange (solid line).

valents of Figs. 1 and 2, respectively, obtained by computer solution of Eqs. (1) with parameter values in the ranges of the experiment. More details appear in the figure captions. The theoretical data also cover another interesting region at a driving frequency about twice that chosen the experiments. Preliminary observations in that frequency range show also a qualitative agreement between the computer solution and the experiments.

We conclude with a new experimental feature whose connection with turbulence has recently been shown.[8] As stated in Ref. 8, $1/f$ type low-frequency divergences, with power spectra as $f^{-\alpha}$ ($\alpha=0.6$–$1.2$), appear when the following conditions are fulfilled: (i) There are at least two basins of attraction; (ii) the attractors have become strange and any random noise (always present in a macroscopic system) acts as a bridge, triggering jumps between them. These jumps have the $f^{-\alpha}$ feature. In the region of bistability (see Fig. 2) we have increased the modulator amplitude $m$ up to the point where the two attractors have become strange. Figure 5 shows the sudden increase in the low-frequency spectrum. The divergent part has a power-law behavior $f^{-\alpha}$ with $\alpha \approx 0.6$. The theory of these new phenomena is under investigation.

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## Correlation Inequalities and Hidden Variables

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A counterexample is given to a recent suggestion that if the inequalities of Clauser and Horne hold, then there exists a hidden-variable model for a spin-$\frac{1}{2}$ correlation experiment of the Einstein-Podolsky-Rosen type.

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Let $p_{ik}(m,m')$ be the distribution for measuring spin components $m$ and $m'$ along directions $\hat{a}_i$ and $\hat{b}_k$ in a two-particle spin-$\frac{1}{2}$ correlation experiment such as Bohm's version of the Einstein-Podolsky-Rosen experiment.[1,2] Consider a model for a set of such distributions associated with different choices for the two directions $\hat{a}_i$ and $\hat{b}_k$, that attempts to represent them all in the form

$$p_{ik}(m,m') = \langle f_i(m,z) g_k(m',z)\rangle , \qquad (1)$$

where $f_i$ and $g_k$ are conditional distributions, one for each particle, and the average is over a distribution of hidden variables $z$ that does not depend on $i$ or $k$.[3] The Clauser-Horne inequalities[4] are a set of necessary conditions that any set of

 