## Deterministic Chaos in Passive Q-Switching Pulsation of a CO<sub>2</sub> Laser with Saturable Absorber

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A single-mode laser with a saturable absorber exhibits spontaneous pulsation in the output (passive Q switching). Period-doubling bifurcation and chaotic passive Q switching which are predicted on the basis of the recently proposed model of the laser system have been observed by use of a CO<sub>2</sub> laser with a formic acid absorber. This is the first experimental evidence of deterministic chaos in a single-mode laser system with a saturable absorber.

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For the past ten years the laser has attracted considerable attention as a test bench for dynamical processes derived from nonlinear equations. Various types of laser systems have been demonstrated experimentally and theoretically to exhibit deterministic chaos. The first observation of optical chaos in laser systems was realized by Arecchi et al.<sup>1</sup> in a CO<sub>2</sub> laser whose cavity loss was modulated by an electro-optical modulator. Some other experiments on CO<sub>2</sub> lasers with electro-optical devices were carried out successively, yielding clear evidence of deterministic chaos.<sup>2,3</sup> The single-mode instability in a homogeneously broadened laser<sup>4</sup> associated with the Lorenz-Haken instability<sup>5</sup> was experimentally obtained in an NH<sub>3</sub> far-infrared laser by Weiss et al.<sup>6</sup> Inhomogeneously broadened gas lasers were theoretically proved to show self-pulsing instability and chaotic behavior in the bad-cavity regime.<sup>7,8</sup> These instabilities were actually observed in He-Ne lasers<sup>9,10</sup> and Xe lasers.<sup>7,11</sup>

On the other hand, it has been well known that the single-mode laser containing a saturable absorber inside its cavity shows periodic self-pulsation called passive Q switching (PQS).<sup>12</sup> Different types of PQS pulse shapes are observed depending on the lasing conditions and characteristics of the absorbers.<sup>13</sup> Recently we have developed an effective rate-equation model for PQS in CO<sub>2</sub> lasers (the three-level:two-level model) where the laser medium is described as a three-level system and the vibrational relaxation of the lower laser level is included as an important process to produce the transient pulse structure.<sup>14,15</sup> All the PQS pulse shapes so far observed in a CO<sub>2</sub> laser were reproduced with detailed fidelity in the computer simulations.<sup>14-16</sup>

The three-level:two-level model predicted for the first time that not only periodic but chaotic PQS pulsation could be realized.<sup>16</sup> According to the rate-equation analysis, the PQS behavior is sensitively dependent on the degree of saturation of the absorption lines. The chaotic pulsation is expected only in limited ranges of the absorber parameters such as an absorption coefficient and a population relaxation rate. These critical conditions of the saturable absorber have been fulfilled, and chaotic PQS has been actually observed in a CO<sub>2</sub> laser. Here we report the first experimental evidence of deterministic chaos in a single-mode laser system with a saturable absorber.



FIG. 1. (a)-(e) PQS-pulse trains and (a')-(e') their power spectra observed as functions of the discharge current *i*. The power spectra are plotted in logarithmic units. The chaotic pulsation [(d)] appears following the period-doubling bifurcation [(a)-(c)]. A periodic window [(e)] appears at 6.3 mA.

The CO<sub>2</sub> laser used in the present experiment consists of a 2.1-m-long gain tube and a 35-cm-long intracavityabsorption cell. The total cavity length is 3.5 m. Gas mixture of CO<sub>2</sub>, N<sub>2</sub>, and He (1:1:8) is flowing through the laser tube at a total pressure of 13.4 Torr. The laser is oscillating in the single longitudinal mode of the 9- $\mu$ m R(20) line. The detailed experimental setup is described in Ref. 14. Formic acid is used as a saturable absorber, and SF<sub>6</sub> is added to the absorber as a buffer gas. The absorption is controlled by change of the pressure of formic acid and by fine adjustment of the laser frequency. A favorable value of the relaxation rate of the absorptive levels is achieved by our changing the pressure of SF<sub>6</sub>.

Figure 1 shows the observed PQS-pulse trains as a function of the discharge current together with their power spectra when the pressures of formic acid and  $SF_6$ are 14 and 374 mTorr, respectively. As the discharge current is increased, period-doubling bifurcation successively occurs. The power spectrum in Fig. 1(a') has a frequency component f and its harmonics. The f/2 and f/4 components and their overtones are clearly observed in the power spectra of period-2 and period-4 time series [see Figs. 1(b') and 1(c')]. The chaotic pulsation has a broad-band spectrum and is shown in Fig. 1(d'). A periodic window is observed at a discharge current of 6.3 mA [Fig. 1(e)]. This window seems to have f/3 and f/6 frequency components although the value of f is shifted to higher frequency because of an increase in the discharge current. At larger discharge current the laser oscillates in the cw mode.

PQS-pulse trains calculated on the basis of the threelevel:two-level model are shown in Fig. 2. The pumping rate is varied in the calculation, corresponding to the discharge current. The formulation of the rate equations and parameter values appears in Ref. 16. Good agreement in the features of the period-doubling bifurcation, chaotic behavior, and the periodic window are obtained between the observation and the calculated results.

In order to make sure that the observed irregular pulsation is really deterministic, we have measured an entropy  $K_2$  which was defined by Grassberger and Procaccia.<sup>17</sup> This quantity  $K_2$  gives a good estimation for a lower bound of the Kolmogorov entropy<sup>18</sup> and has the following properties: (1)  $K_2$  is infinite in a random non-



FIG. 2. (a)-(e) PQS-pulse trains and (a')-(e') their power spectra as functions of the pumping rate P obtained from numerical calculations based on the three-level:two level rate-equation model. The power spectra are plotted in logarithmic units.

deterministic system; (2)  $K_2$  is positive constant in a chaotic system; and (3)  $K_2=0$  in an ordered system. The procedure introduced by Grassberger and Procaccia is applied to the experimentally obtained digitized time series of the laser output  $\{X_i\}_{i=1}^N$ .  $X_i$  is normalized to a typical value at the pulse peak of the time series in Fig. 1(a). The entropy  $K_2$  is calculated as follows:

$$C_d(\epsilon) = \lim_{N \to \infty} N^{-2} \left[ \text{number of pairs } (n,m) \text{ with } \left[ \sum_{i=1}^d |X_{n+i} - X_{m+i}|^2 \right]^{1/2} < \epsilon \right], \tag{1}$$

$$K_{2} = \lim_{\substack{d \to \infty \\ \epsilon \to 0}} K_{2,d}(\epsilon) = \lim_{\substack{d \to \infty \\ \epsilon \to 0}} \tau^{-1} \ln[C_{d}(\epsilon)/C_{d+1}(\epsilon)].$$
(2)

Here  $C_d(\epsilon)$  is the correlation integral at the embedding dimension d, and  $\tau$  is the sampling time of the time series.

Figure 3(a) shows  $K_{2,d}$  for the irregular pulsation in Fig. 1(d) as a function of d. For comparison,  $K_{2,d}$  for the periodic case of Fig. 1(a) is plotted in Fig. 3(b). The value of  $K_{2,d}$  seems to approach a finite constant in Fig. 3(a),



FIG. 3. The entropy  $K_{2,d}$  of the observed data sets plotted as a function of the embedding dimension d for different values of  $\epsilon$  (a) in the case of the irregular pulsation of Fig. 1(d); (b) in the case of the periodic pulsation of Fig. 1(a).

suggesting that the data set is taken from a chaotic orbit. On the other hand,  $K_{2,d}$  for the periodic signal tends to progress toward zero although its convergence is somewhat restricted by the noise superposed on the periodic time series.

In conclusion, deterministic chaos has been experimentally demonstrated to appear in the passive Q-switching pulsation of a CO<sub>2</sub> laser with a formic acid absorber. The single-mode instabilities so far observed in the intrinsic (without any electric devices) laser system<sup>6,7,11</sup> are qualitatively interpreted to be caused by the modesplitting effect due to strong dispersion. These instabilities are described by the Maxwell-Bloch equations which include the molecular polarizations and the phase of the electromagnetic field.<sup>4,7,8</sup> On the other hand, the present instability is well reproduced through numerical analysis based on the rate equations. This means that the present chaos is caused only by the transient variations of the populations of the relevant energy levels in the amplifier and absorber. The lasers which exhibit chaos in the single-mode oscillation have been limited to high-gain lasers such as He-Ne lasers and Xe lasers since the bad-cavity condition is generally needed.<sup>4,5,7,8</sup> The present experiment has extended the range of possibility to observe deterministic chaos in laser systems, showing that a saturable absorber inside the laser cavity can provoke chaotic passive Q switching in a small-gain laser far from the badcavity condition.

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