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Instability and chaos in two-mode oscillation of a CO_2 laser modulated by a saturable absorber

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Two-mode instability in a CO_2 laser with a saturable absorber is investigated both experimentally and theoretically. A strong correlation is found between the mode competition and the passive Q-switching behavior. The observed two-mode instability is nicely reproduced by our rate-equation model in which the vibrational relaxation processes and the spatial mode coupling are considered. Furthermore, numerical calculation predicts existence of chaos in the two-mode oscillation with a higher fractal dimension than the single-mode chaos.

Recently, a laser with a saturable absorber (LSA) has attracted considerable interest as a useful model to exhibit fundamental aspects of nonlinear dynamics, such as limit cycles, chaos, and bifurcations.[1] The passive Q-switching (PQS) instabilities in the LSA are critically dependent on the energy-level structure and dynamic properties of the gain and loss media. This also makes it possible to use the laser dynamics as a probe of molecular processes.[2]

So far, most of the studies on the LSA dynamics have been focused on the single-mode characteristics.[3] Several types of regular PQS pulsations were observed in single-mode CO_2 and N_2O lasers; a spikelike pulse, sinusoidally modulated output, and a pulse with undamped undulation on its tail.[4,5] Deterministic chaos and Feigenbaum's scenario also appeared in certain parameter regions.[6–8] Recently, we developed a rate-equation model (the three-level–two-level model) for the single-mode CO_2 and N_2O lasers.[2,9] Computer simulations based on the three-level–two-level model (the 3-2 model) successfully reproduced the observed regular and chaotic PQS pulsations and their dependence on laser parameters.[10]

Introduction of an extra lasing mode, which is nonlinearly coupled to the main mode, may increase the phase-space dimension and enrich the LSA dynamics.[11] In this paper, we report a new type of instability performed by two lasing modes of a CO_2 LSA, one of which is passively Q switched by the saturable absorber.[12] The 3-2 model, modified to fit the multimode case, is applied to analyze the observed characteristics of mode competition.

An experimental setup and procedures are described in detail elsewhere.[2] Figures 1(a)–1(c) show observed temporal behavior of the two-mode oscillation. The laser oscillation occurs on two lines which are in axial (TEM_{00}) and off-axial (TEM_{01}) modes of the Fabry-Pérot cavity. The laser line in the axial mode is modulated by passive Q switching. The feature of mode competition is critically dependent on the PQS behavior of the axial mode. When a PQS pulse with undamped undulation appears in the axial mode, the laser oscillation is alternately switched from one mode to the other mode, as is shown in Fig. 1(a). On the other hand, both modes simultaneously oscillate when the axial mode is sinusoidally modulated through PQS [see Figs. 1(b) and 1(c)]. In the case of Fig. 1(b), the intensities of the two modes change out of phase simultaneously. A small change in the grating angle results in the in-phase intensity modulation as shown in Fig. 1(c). In Figs. 1(a) and 1(b) relaxation oscillation is observed in the off-axial mode, superposed on the quasi-cw tail and the sinusoidal modulation, respectively.

The competition between the axial and off-axial modes comes from cross saturation caused by spatial overlapping between the two modes. By considering this effect, we modify the 3-2 model to reproduce the observed two-mode instability. Figure 2 shows a schematic diagram of the two-mode 3-2 model. As is shown in Fig. 2(a), the cylindrical laser gain medium is considered to consist of two regions (the center region and the side region). In the center region, the TEM_{00} mode saturates the laser gain more strongly than the TEM_{01} mode, and vice versa, in the side region. The gain medium in each region is represented by three vibrational levels, the upper laser level (001), the lower laser level (100) or (020), and the ground level [see Fig. 2(b)]. The vibrational relaxations are introduced among the three levels. The absorption medium is represented simply by two rotation-vibration levels in resonance with the axial mode laser radiation. The populations in the two levels are assumed to relax to thermal equilibrium values at the same rate.

Based on the model, the dynamics of the laser system is described by seven nonlinearly coupled rate equations as follows:

$$\frac{dn}{dt}=B_g f_g[(M_{c1}-M_{c2})+C_1(M_{s1}-M_{s2})]n l_g/L-(B_a N l_a/L+k)n \ , \quad (1)$$

$$\frac{dn'}{dt}=B'_g f'_g[(M_{s1}-M_{s2})+C_2(M_{c1}-M_{c2})]n' l_g/L-k'n' \ , \quad (2)$$

$$\frac{dM_{c1}}{dt}=-(B_g f_g n+C_2 B_g' f_g' n')(M_{c1}-M_{c2})+P_c(M_c-M_{c1}-M_{c2})-(R_{10}+R_{12})M_{c1}+R_m(M_{s1}-M_{c1}) , \tag{3}$$

$$\frac{dM_{c2}}{dt}=(B_g f_g n+C_2 B_g' f_g' n')(M_{c1}-M_{c2})+R_{12}M_{c1}-R_{20}M_{c2}+R_m(M_{s2}-M_{c2}) , \tag{4}$$

$$\frac{dM_{s1}}{dt}=-(B_g' f_g' n'+C_1 B_g f_g n)(M_{s1}-M_{s2})+P_s(M_s-M_{s1}-M_{s2})-(R_{10}+R_{12})M_{s1}+R_m(M_{c1}-M_{s1}) , \tag{5}$$

$$\frac{dM_{s2}}{dt}=(B_g' f_g' n'+C_1 B_g f_g n)(M_{s1}-M_{s2})+R_{12}M_{s1}-R_{20}M_{s2}+R_m(M_{c2}-M_{s2}) , \tag{6}$$

$$\frac{dN}{dt}=-2B_a nN-r(N-N^*) . \tag{7}$$

Here n (n') is the mean photon density of the TEM_{00} (TEM_{01}) mode over the center (side) region. M_{c1} (M_{s1}) and M_{c2} (M_{s2}) are, respectively, the population densities of the upper and lower laser levels in the center (side) region. N is the population density difference between the absorptive levels in the absorber. The coupling constants C_1 and C_2 are determined by the transverse intensity distribution of the TEM_{00} and TEM_{01} modes. R_m is the rate constant for diffusion of CO_2 molecules between the center and side regions. Notations of the other parameters are identical with the single-mode 3-2 model (see Ref. 9), although the subscripts c and s, respectively, indicate the parameters in the center and side regions, and a prime indicates parameters referring to the off-axial mode.

Figures 1(d)–1(f) show results of numerical integration of the rate equations with parameter values reasonably adjusted for the real CO_2 laser system. The calculation reproduces well the detailed characteristics of the observed mode competition, such as PQS pulse shapes in the axial mode, relaxation oscillation in the off-axial mode, and the switching frequencies. The numerical analysis reveals that in the case of Fig. 1(d), the gain for the off-axial mode is strongly saturated by the first high peak at the onset of a PQS pulse. Eventually, the off-axial mode stays off until the PQS pulse is terminated. In

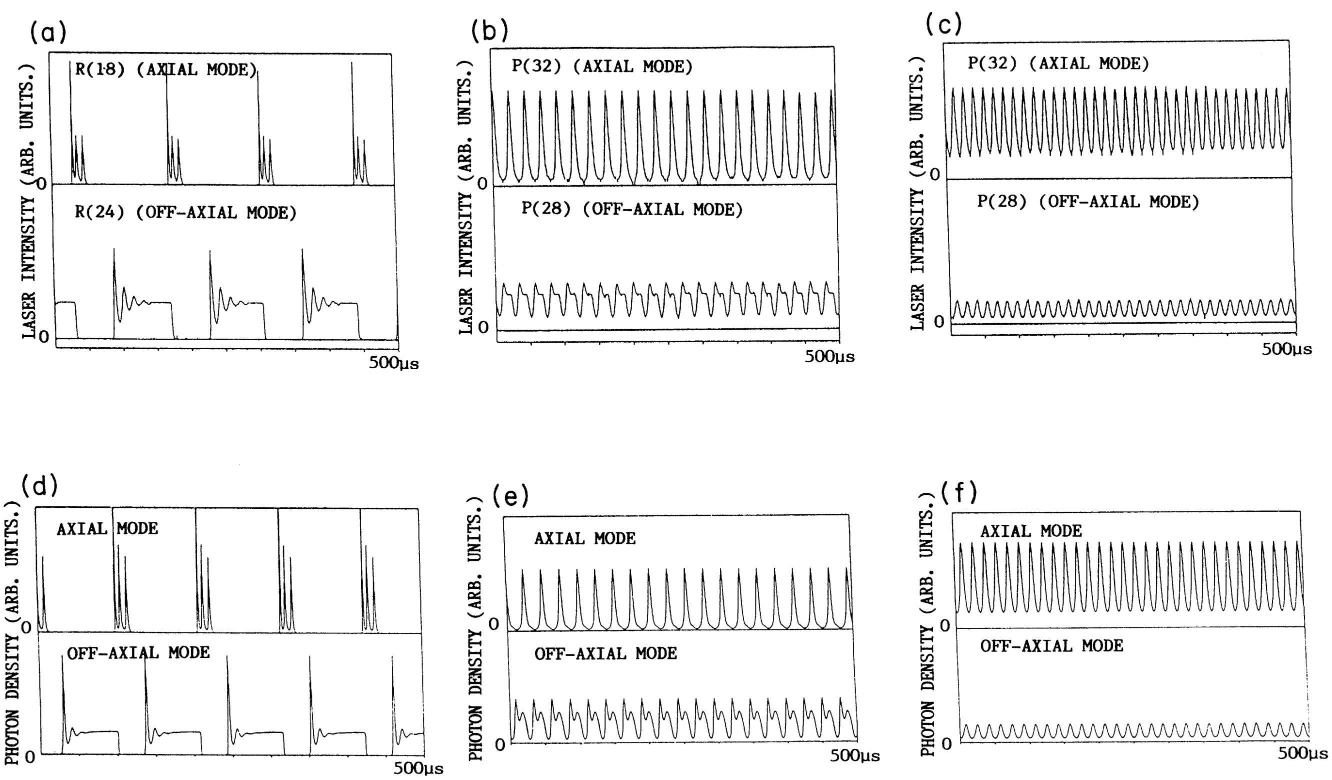

FIG. 1. Observed [(a)–(c)] and calculated [(d)–(f)] periodic two-mode competition. (a) The 9-μm $R(18)$ line is passively Q switched by the HCOOH absorber (290 mTorr). (b) and (c) The 9-μm $P(32)$ line is modulated by the CH_3OH absorber (850 mTorr). The grating angle is slightly different between (b) and (c).

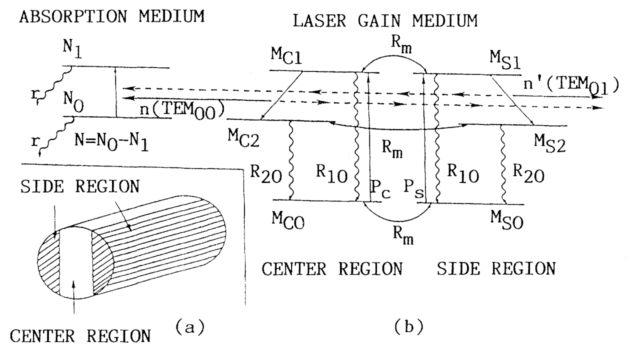

FIG. 2. (a) Two regions in the laser gain medium. (b) Schematic representation of the 3-2 model for the two-mode oscillation. The photon densities of the TEM_{00} and TEM_{01} modes are denoted by n and n', respectively. The population densities in the gain and loss media are represented by M and N, respectively. Detailed notations are described in the text.

the case of Figs. 1(e) and 1(f), on the other hand, the peak intensity of the sinusoidal PQS in the axial mode is not large enough to reduce the off-axial mode gain far below the laser threshold.

According to the rate-equation analysis, the parameter regions for the types of mode competition are investigated and demonstrated in the phase diagram in Fig. 3. The phase diagram is depicted for the pumping rate $P_c(=P_s)$

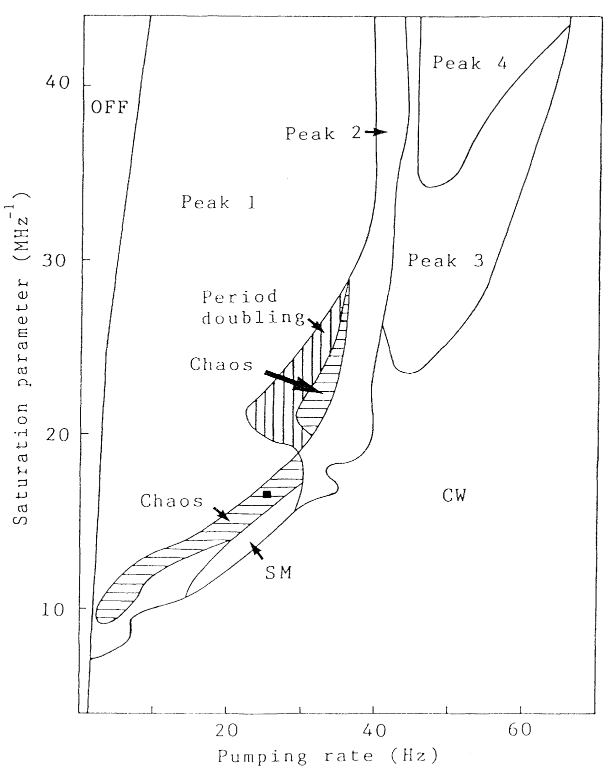

FIG. 3. Theoretically obtained phase diagram for the pumping rate and the saturation parameter. The square in the chaos region indicates the parameter values at which the strange attractor of Figs. 4(b) and 4(d) appears.

in the gain medium and the saturation parameter in the absorption medium. The saturation parameter in this case is proportional to the absorption cross section divided by the population relaxation rate.[10] The behavior of the mode competition can be classified depending on the PQS pulse shape in the axial mode. Peak i denotes the region where the axial mode shows a regular pulse train. i indicates the number of peaks involved in a single pulse. The exclusive mode exchange can be realized in these regions [the time series of Fig. 1(d) appears in the peak-3 region, for example]. SM denotes the region where the axial mode intensity is sinusoidally modulated. The simultaneous two-mode oscillation appears in this region. Laser oscillation in both modes is cw in the region denoted by CW. Only the off-axial cw mode oscillation is realized in the region denoted by OFF.

There are two regions where chaotic two-mode oscillation is observed. In the region located on the upper side, chaos appears after the period-doubling bifurcation when the pumping rate is increased. Quasiperiodic and subharmonic pulsations are found in the vicinity of the lower-side region for chaos. However, no clear standard scenario to reach chaos has not yet been identified at the present stage.

Here we compare the two-mode chaos with the single-mode chaos calculated from the 3-2 model.[8,13] Figures 4(a) and 4(b) show the return maps (the Lorenz plot)[14] of the axial mode intensity constructed from the chaotic time series. The two-mode chaos is calculated at the parameter values indicated in the phase diagram. The return map of the single-mode chaos traces a one-dimensional unimodal curve. The mechanism to produce the chaos is well interpreted from the Feigenbaum's scenario.[15] On the other hand, the Lorenz plot of the

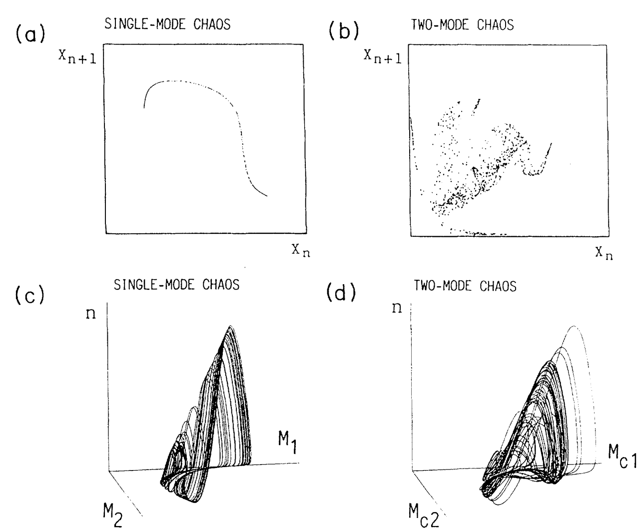

FIG. 4. Return maps of the photon density in the axial mode [(a) and (b)] and phase portraits of the strange attractors [(c) and (d)]. In the return maps, the $(n+1)$th peak value X_{n+1} of the axial-mode photon density is plotted as a function of the nth peak value X_n. The phase space is defined for the photon density n, the upper-laser level population density M_1 (M_{c1}), and the lower-laser level population density M_2 (M_{c2}).

two-mode chaos shows two-dimensional distribution, suggesting that its strange attractor has a higher dimension than the single-mode case. Figures 4(c) and 4(d) show portraits of the strange attractors in the phase space for the axial-mode intensity and the population densities in the upper and lower laser levels. The attractor of the single-mode chaos has a nearly two-dimensional structure, while that of the two-mode chaos is distributed three dimensionally. The correlation dimension[16] of the attractors is measured to be 2.01 ± 0.01 and 2.47 ± 0.01 for the single- and two-mode chaos, respectively. When the variables related to the off-axial mode quickly follow the changes in the axial-mode variables and their behavior gives no effect to the laser dynamics, the fractal dimension of the two-mode strange attractor remains the same as the single-mode chaos. A difference in the fractal dimensionality implies that the laser dynamics is appreciably modified by the cross saturation between the two modes, leading to the appearance of the new type of chaos. Experiments to search for the two-mode chaos are now in progress by use of a CO_2 laser.

The two-mode instability in a CO_2 LSA may have an analogy with the mode hopping and mode partition generally observed in diode lasers.[17] Although the former is deterministically driven by PQS and the latter are triggered by the spontaneous emission, both dynamics originate from the cross saturation of the laser gain. It is important in analyzing the diode laser instability to describe detailed carrier dynamics. The present rate-equation analysis may help to understand the mechanism of the dynamic mode competition in diode laser systems.

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