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## Observation of bifurcation to chaos in a passive all-optical Fabry–Perot resonator: a high-frequency optical modulator

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We report the first observations of bifurcation routes to chaos in an all-optical resonator. Generation of associated deep and sustained Ikeda oscillation of the smooth CO<sub>2</sub> laser input pulses at twice the round-trip time of the Fabry–Perot resonator provides a high-frequency (*ca.* 0.1 GHz) passive optical modulator device. Results are in excellent agreement with our adaption of optical bistability theory to the time-dependent régime.

The transition to chaos is of current interest throughout physics. Among systems exhibiting chaos, ‘optically bistable’ devices merit special attention as basically simple systems (Ikeda 1979; Nakatsuka *et al.* 1983). Of these a two level medium within an optical resonator is the most fundamental and furthermore is fully quantizable (Harrison *et al.* 1983). We report here first experimental evidence for chaotic behaviour in such a system. NH<sub>3</sub> gas is used as the nonlinear medium contained in a Fabry–Perot resonator optically pumped by the smooth pulses from a CO<sub>2</sub> laser. Deep Ikeda oscillation at twice the ‘round-trip’ time ( $2t_r$ ) is obtained and, by varying the parameters, we observe  $\frac{2}{3}t_r$  modulation, and period-doubling to  $4t_r$  and  $\frac{4}{3}t_r$ . These phenomena, coupled with theory, suggest that the aperiodic pulses that we also observe represent the first evidence for all-optical chaos in a quantum system.

We also report first observations of optical hysteresis in this system by using SF<sub>6</sub>, a precursor to possible generation of nonlinear instabilities under (continuous wave) (c.w.) conditions in this and similar molecules with low saturation intensities.

The Fabry–Perot cavities containing ammonia gas at *ca.* 5–40 Torr were pumped by smooth TEA CO<sub>2</sub> laser pulses of *ca.* 100 ns duration and peak intensity *ca.* 10–20 MW cm<sup>-2</sup>. Oscillation was observed by using a number of laser transitions near-coincident with NH<sub>3</sub> absorption lines. Here we concentrate on the 10R(14) CO<sub>2</sub> line, pumping the aR(1,1) NH<sub>3</sub> transition 1.23 GHz above line centre (Garing *et al.* 1959). This transition is pressure broadened above *ca.* 5 Torr (27 MHz Torr<sup>-1</sup>† at f.w.h.m.), where it acts as a homogeneously broadened two-level system pumped off resonance (Harrison *et al.* 1984a).

The resonators of length 40–100 cm comprised a single-surface Ge flat input coupler of reflectivity  $R_0 = 36\text{--}85\%$  and a single surface Ge output coupler, of 2 m radius of curvature and reflectivity  $R_L = 76\%$ . The input and output signals were monitored by photon-drag detectors and a Tektronix model 7104 oscilloscope; total resolution  $\lesssim 1$  ns. The output coupler was equipped with PZT tuning encompassing one free spectral range of the Fabry–Perot resonator. The transverse intensity profile of the input signal was Gaussian with a 1/e spot diameter of *ca.* 3 mm. An optical delay line prevented feedback of the smooth 100 ns (f.w.h.) CO<sub>2</sub> laser pulses from the nonlinear resonator to the laser.

† 1 Torr  $\approx$  133.322 Pa.

Representative examples of the modulated output for PZT tuning are shown in figure 1*a* for cavity length 86 cm, cell length 70 cm and pressure 10 Torr, with input coupler reflectivity 67%. Strong Ikeda oscillation (period *ca.* 13 ns, close to  $2t_r = 11.5$  ns), persistent throughout the pulse, is evident in the neighbourhood of minimum transmission, consistent with the four-wave mixing interpretation of this instability (Firth 1981; Firth *et al.* 1982). (Note that in contrast to the ring resonator the Fabry–Perot geometry does not prescribe  $2t_r$  as the basic period for Ikeda oscillation.) PZT tuning of the cavity leads progressively to ‘switching’ behaviour with high peak transmission followed by damped oscillation of longer period.

At lower pressures (4–8 Torr), where inhomogeneous broadening may be important, we also obtained strong and sustained  $4t_r$  oscillation. At higher pressures (20–30 Torr) much more complex pulse shapes were obtained. These features were enhanced for reduced input coupler reflectivity ( $R_0 = 36\%$ ), since large input coupling is needed to bleach the high absorption

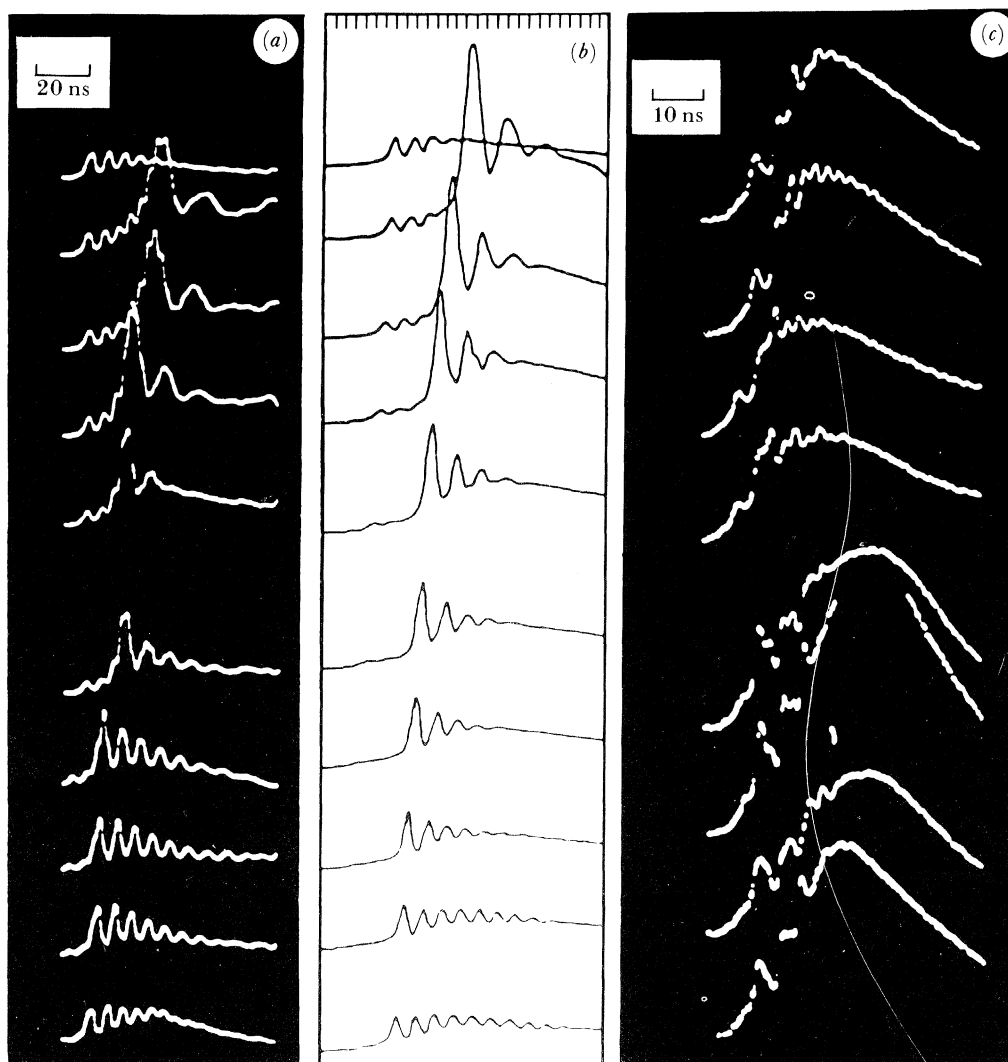


FIGURE 1. (a) PZT scan showing Ikeda oscillation at 10 Torr; cavity length = 86 cm;  $R_0 = 67\%$ ;  $R_L = 76\%$ .  $\text{NH}_3$  transition  $aR(1, 1)$ . (b) Computed traces for parameters given for (a);  $\alpha l = 1.75$ ,  $\Delta = 10$ ; one scale division is equivalent to  $t_r$  (cavity round-trip time). (c) PZT scan showing  $2t_r$ ,  $\frac{3}{2}t_r$ ,  $3t_r$  modulation and aperiodic pulses at 19 Torr. Cavity parameters as given for (a) with  $R_0 = 36\%$ .

( $\alpha L = 6$ ) to achieve adequate cavity feedback. The PZT sequence shown in figure 1*c* for a pressure of 19 Torr shows  $2t_r$  oscillation (top trace), developing to  $\frac{3}{2}t_r$  oscillation on the higher branch, bifurcating to  $\frac{4}{3}t_r$  before again evolving to lower-branch  $2t_r$  modulation (bottom trace). Aperiodic pulse shapes, characteristic of chaos, are also evident here and in other data taken at similar pressures.

Observation of these effects under c.w. conditions requires molecules with low saturation intensities, such as SF<sub>6</sub> (*ca.* 6 W cm<sup>-2</sup> Torr<sup>-1</sup>). As a precursor to this we report observation of optical hysteresis effects in this gas over a wide range of parameter conditions. We note that operation here is in the ‘bad cavity’ limit and so precludes generation of Ikeda instabilities. Fabry–Perot resonators, ranging in length from 2 to 15 cm, with intra-cavity gas cells as short as 1 mm were operated at SF<sub>6</sub> gas pressures ranging from *ca.* 1 to *ca.* 200 Torr. Various lines in the 10P band of the TEA CO<sub>2</sub> laser were used to pump the dense and broad spectral features of the  $\nu_3$  vibrational mode of SF<sub>6</sub> near resonance (Harrison & Butcher 1980). Effects of switching, power limiting, and overshoot with nanosecond response times, limited only by cavity decay time, were routinely obtained at gas pressures commensurate with self focusing. Sample traces of the transmitted signal for PZT tuning the cavity through half a spectral range are shown in figure 2*b*, cavity length 3 cm, cell length 1 cm with SF<sub>6</sub> at a pressure of *ca.* 10 Torr pumped at 947.7 cm<sup>-1</sup> (10P16 CO<sub>2</sub> line). Corresponding recordings of instantaneous input (*x*-axis) and output (*y*-axis) intensity (figure 2*c*) show pronounced optical hysteresis effects. Equivalent recordings for the empty cavity are shown in figure 2*a* for reference.

Stability analysis of nonlinear Fabry–Perot resonators has apparently only been examined in the dispersive (Kerr) limit (Firth 1981). Our experiments, however, necessarily involve the bleaching of a rather substantial absorption. We have therefore adapted the model of Carmichael & Hermann (1980), which treats steady-state optical bistability in a Fabry–Perot resonator, handling the time dependence by replacing the gas cell by *N* thin slices symmetrically placed within the cavity. Assuming the adiabatic limit, we apply steady-state theory to find the forward and backward transmissions of each slice (unequal due to the phase–population grating). We can then follow the field around the cavity, where it interacts in the slices with *N* earlier and *N* later fields, and keep track of its attenuation at each stage. At the output and input couplers we apply the usual Fabry–Perot boundary conditions, and thus have, in effect, a  $2(N+1)$  parameter mapping problem. Full details of this analysis will be reported elsewhere (Harrison *et al.* 1984*b*).

Application of this procedure to NH<sub>3</sub> and use of an input pulse of the form shown in figure 2*a* yields the transmitted pulse shapes in figure 1*b*, in pleasing agreement with the observed pulse shapes, especially since only measured parameters are used:  $\alpha$  is 0.025 cm<sup>-1</sup> at 10 Torr and scales as  $p^2$  (this, plus the pressure broadening rate and constants gives a saturation intensity  $I_s \approx 2.3$  MW cm<sup>-2</sup> at 10 Torr (2.5 MW cm<sup>-2</sup> at 30 Torr)). The value  $I/I_s = 7$  is thus in line with the measured input intensities in the range 10–20 MW cm<sup>-2</sup>.

This good agreement is rather surprising in view of the omission of reservoir (Harrison *et al.* 1983, 1984*a, b*) and transverse effects in the analysis. Inclusion of such effects, in particular transverse effects (Moloney *et al.* 1982), will, however, be necessary in predicting the more complex pulse shapes of figure 1*c*, though qualitative agreement still exists from the plane-wave approach. It is emphasized that for c.w. inputs these models give steady-state oscillation and chaos within our experimental parameter range and we have identified period doubling to  $16t_r$  en route to chaos in one case.

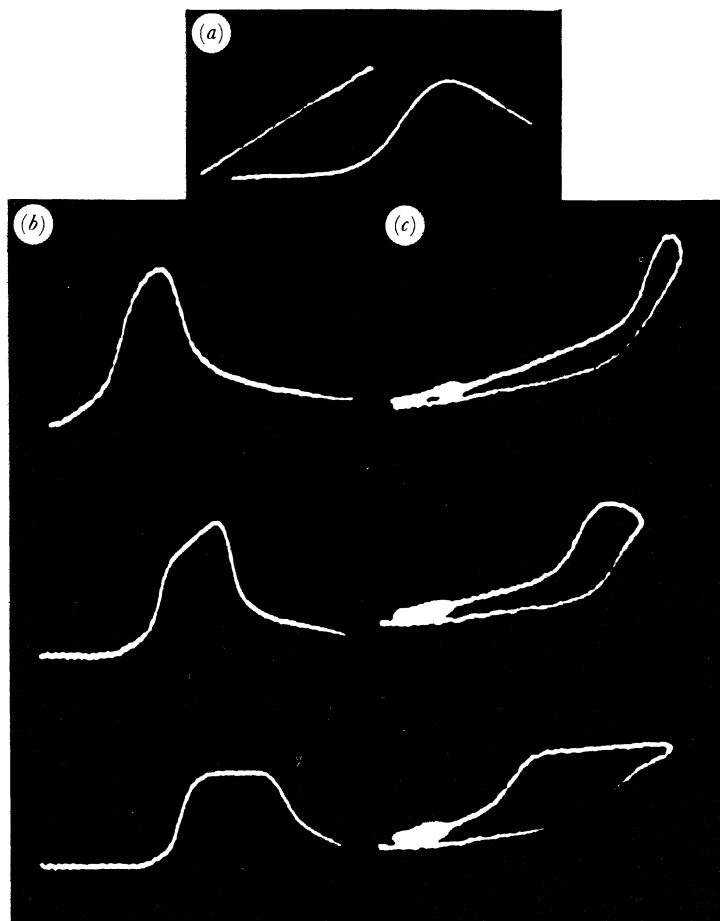


FIGURE 2. (a) Typical single-mode input pulse together with  $x$ - $y$  trace of instantaneous input and output intensity for empty Fabry-Perot resonator. (b) PZT scan (over half of the free spectral range) of transmitted signal, SF<sub>6</sub>, pressure cavity length 3 cm, cell length 1 cm. (c) Corresponding  $x$ - $y$  traces of input and transmitted signal intensity.

In conclusion, our observations of  $2t_r$  oscillation period doubling to  $4t_r$  and, over a more limited parameter range,  $\frac{2}{3}t_r$ ,  $\frac{4}{3}t_r$  modulation and aperiodic pulse shapes in NH<sub>3</sub> gas confirm the predicted behaviour of this system and support the conclusion that we have driven an all-optical two-level system through oscillation to chaos. Already of use as a passive optical modulator ( $2t_r \approx 100$  MHz) of pulsed signals, prospects for modulation in the gigahertz range are promising. The possibility of generating these effects under c.w. conditions is supported by our observations of strong optical hysteresis in this system by using SF<sub>6</sub> which, along with many other molecular gases, exhibits low saturation intensity.

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