Observation of Period Doubling in an All-Optical Resonator Containing NH_3 Gas

R. G. Harrison, W. J. Firth, C. A. Emshary, and I. A. Al-Saidi
Department of Physics, Heriot-Watt University, Riccarton, Edinburgh EH14 4AS, United Kingdom
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Two-round-trip-time modulation of the cavity field has been observed in a passive ring resonator, containing an ammonia cell, pumped by smooth 100-ns CO_2 laser pulses. The results are in excellent accord with the theory of Ikeda instability instability in a two-level system, which is generalized to include reservoir kinetics.

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Systems exhibiting period-doubling cascades to chaotic behavior while governed by deterministic equations have attracted great interest.[1,2] Passive all-optical systems are particularly interesting here, as basically simple systems capable of exhibiting oscillation[3,4] and turbulence, but also because they can be fully quantized. Ikeda[5] showed in 1979 that an optically bistable ring resonator containing a two-level system can show a period-doubling cascade, a sufficiently strong cw input beam yielding an output oscillating at *twice* the resonator round-trip time t_R. On further increase of the input field the output period doubles to chaos. Since then, observations of these phenomena have been made in various optical systems, such as a hybrid bistable device[6] and lasers,[7,8] but the nearest approach to Ikeda's system has been a recent demonstration[9] in a fiber-optic resonator, using mode-locked excitation to avoid stimulated scattering. None of these systems are particularly simple, nor do they lend themselves to quantization. We believe that molecular gases, excited close to resonance by a CO_2 laser, have unique advantages in this field, and here we report observations of $2t_R$ oscillation (with some indications of $4t_R$) in an all-optical system very similar to Ikeda's original proposal.

A passive ring resonator was pumped by a transversely excited atmosphere (TEA) CO_2 laser pulse [$10R(14)$ transition, $\lambda = 10.3\ \mu\text{m}$]. This line lies 1.23 GHz above the $aR(11)$ transition[10] of the NH_3 gas contained in a 1-m intracavity cell at pressures of 9–15 Torr, where it acts as a *homogeneously broadened* two-level system.

The arrangement is illustrated in Fig. 1. The CO_2 hybrid TEA laser/amplifier system yields smooth, single, transverse and longitudinal mode pulses of full width at half maximum ~ 100 ns [Fig. 2(a)] and peak power ~ 1 MW. The laser pulses are coupled, with use of a single-surface Ge flat, $R = 36\%$, into a 3.5-m three-element ring cavity, closed by 100% gold mirrors, containing the gas cell. The input and cavity signals were sampled by KBr beam splitters, and monitored

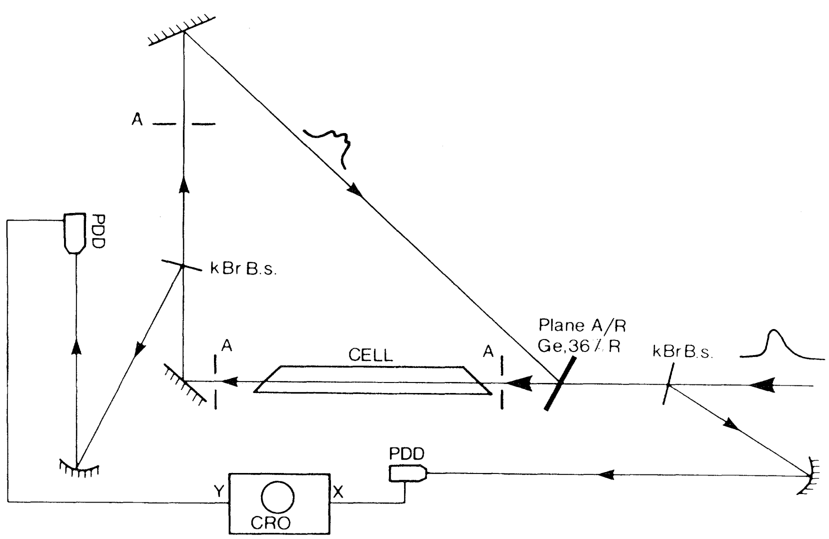

FIG. 1. Schematic diagram of ring-cavity system. B.s.: beam splitter; PDD: photon-drag detector; A/R: antireflection coated.

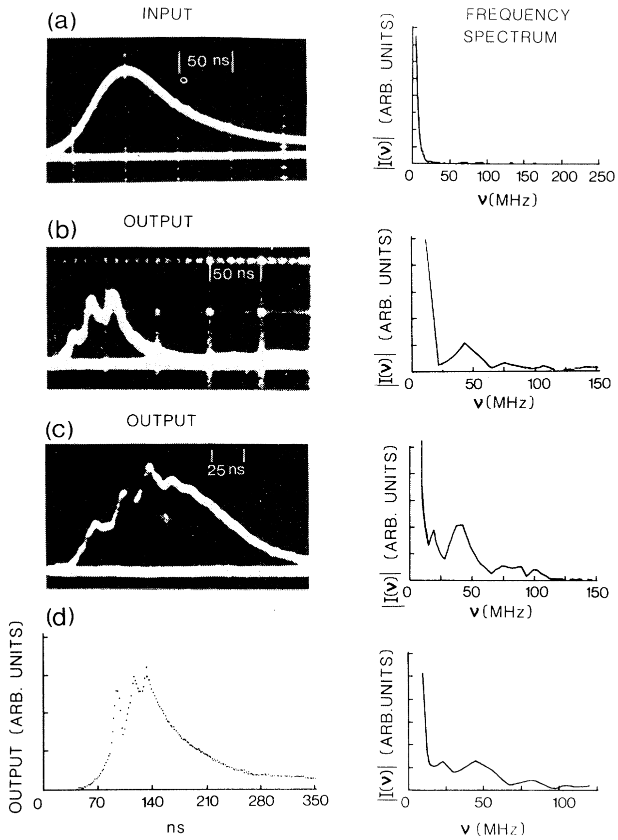

FIG. 2. Sample oscilloscope traces of (a) the pump signal and (b)–(d) the ring-cavity signal, together with their frequency spectra; obtained from digitized traces as shown in (d).

by photon-drag detectors and a Tektronix model 7104 oscilloscope; total response time was $\lesssim 1$ ns. For NH_3 pressures ~9–15 Torr, significant self-focusing was observed in single-pass experiments, confirming a nonlinear refractive-index contribution substantial enough for dispersive optical bistability and Ikeda instability. Closing the ring caused a huge distortion of the pulse shapes (sampled after the NH_3 cell). In particular a considerable proportion of these showed modulation at the 23.4-ns period expected for Ikeda oscillation in our system.

Figure 2 shows representative examples of these modulated pulses. To confirm the period, we have digitized and Fourier transformed the traces; the resulting spectra show pronounced peaks at ~45 MHz, confirming our observation of Ikeda instability: We even have, in two of the cases, a subsidiary peak at $(4t_R)^{-1}$, possibly indicating a further bifurcation. In contrast, the input pulse has an essentially featureless spectrum [Fig. 2(a)].

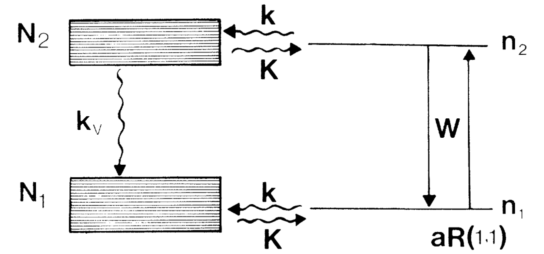

FIG. 3. Kinetic model for NH_3 absorption. Radiative coupling is represented by continuous lines and collisional relaxation (both rotational and vibrational) by wavy lines.

Our system is not quite a two-level system: Allowance must be made for population transfer within the rotational manifolds, as described in detail elsewhere.[11] Here we give a simplified version which demonstrates how the full system may be handled in the context of a passive ring resonator, thereby obtaining a considerable generalization of Ikeda's model.[5]

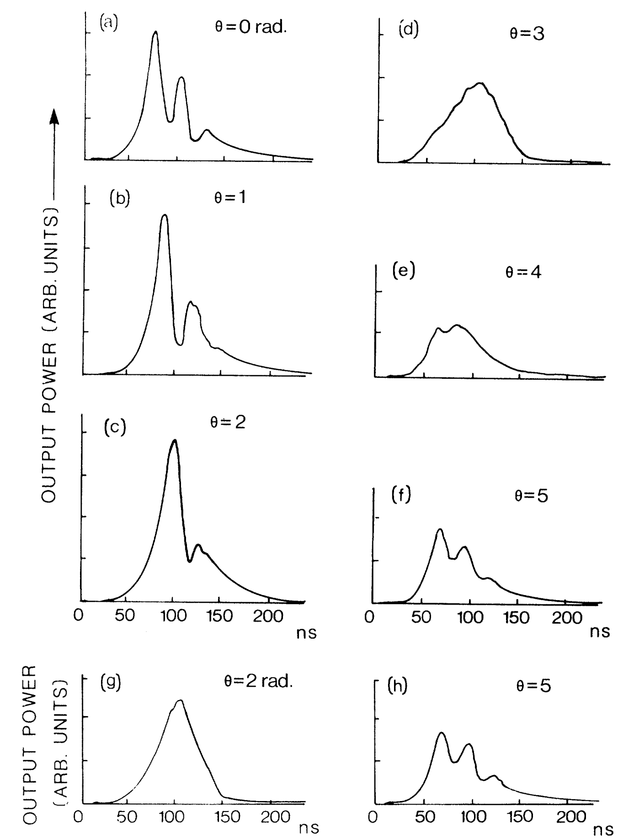

FIG. 4. Numerically determined ring-cavity signals for various cavity tunings using Fig. 2(a) as the pump signal. ($\alpha L = 3$, $kt_R = 5$, $Kt_R = 0.1$, $|\epsilon|_{max}^2 = 0.9$.)

The level scheme is diagrammed in Fig. 3, and leads to the set of rate equations

$$\dot{n}_1 = -W[n_1 - (g_1/g_2)n_2] + KN_1 - kn_1, \quad (1)$$

$$\dot{N}_1 = -KN_1 + kn_1 + k_v N_2, \quad (2)$$

with similar equations for n_2 and N_2. For simplicity, we assume equal level-manifold rate constants in the two levels; g_1/g_2 is the degeneracy factor (here $\frac{5}{3}$) and k_v the vibrational-translational relaxation rate, which is negligibly small for our conditions. These equations conserve total population, and $n_1 + n_2 = N_e$ and $N_1 + N_2 = N_e$ separately (e relating to thermal equilibrium); generalization is straightforward. Detailed balance requires that $kn_e = KN_e$; in the infinite-reservoir limit K thus goes to zero and the system has effectively just two levels. In NH_3, however, $n_e/N_e \sim 2\%$ and $K \sim 2$ μs^{-1} $Torr^{-1}$,[12] so that $kt_R < 1$, as required for Ikeda instability, but $Kt_R > 1$. The major effect of finite K is a leaching of population on a time scale K^{-1}, which eventually kills the instability: Note that absence of oscillation on the falling edge is a feature of our data (Fig. 2).

To convert the rate equations from local equations to take account of propagation and feedback effects, we perform a retarded-time integration,[5] to obtain

$$\epsilon(t) = \epsilon_i + R\epsilon(t - t_R)e^{-i\theta} \exp[-(\alpha L/2)(1 - i\Delta)D(t - t_R)], \quad (3)$$

where $\epsilon(t)$ is the intracavity field and ϵ_i the transmitted incident field, just before the gas cell, both normalized to the saturation field[13]; R and θ are the empty-cavity amplitude loss and phase shift per round trip; α is the small-signal absorption coefficient; Δ is the molecular detuning, $\Delta = (\omega_{CO_2} - \omega_{NH_3})/\gamma$; and (with the assumption $n_{2e} = N_{2e} = 0$)

$$D(t) = (n_e L)^{-1} \int_0^L dz [n_1(z,t') - (g_1/g_2) n_2(z,t')]_{t' = t - (L-z)/c} \equiv \langle n_1 - (g_1/g_2) n_2 \rangle / n_e. \quad (4)$$

The population rate equations become

$$\dot{D} = k\left[\left(1 + \frac{g_1}{g_2}\right)\frac{N_1}{N_e} - \frac{g_1}{g_2} - D(t)\right] - \frac{k}{2}\left(1 + \frac{g_1}{g_2}\right)|\epsilon(t)|^2 \frac{(1 - e^{-\alpha L D(t)})}{\alpha L}. \quad (5)$$

Equations (1) and (2) are essentially unchanged, but the populations are now assumed averaged as in (4).

We have numerically integrated these equations, using the pump pulse of Fig. 2(a) as input, and Fig. 4 shows the predicted intracavity pulses as a function of cavity tuning for representative parameter values. Oscillation at $2t_R$ is manifest in three of these traces, while strong pulse distortion indicative of optical bistability occurs at the opposite tuning, as expected.[13,14] In view of the fact that our present model neglects self-focusing this range of behaviors matches extremely well with the pulse shapes we observe (Figs. 2 and 5). (Because our cavity was free-standing, the value of θ inevitably drifted from shot to shot, so that we sampled the full range of possible pulse shapes.) For comparison of our model with a pure two-level scheme we show two traces [Figs. 4(g) and 4(h)] with K set equal to zero. From comparison with Figs. 4(c) and 4(f) the Ikeda oscillation is essentially unaffected, while the bistable pulse shape is considerably changed—this is not unexpected, since the latter is a first-order transition, whereas Ikeda instability is sec-

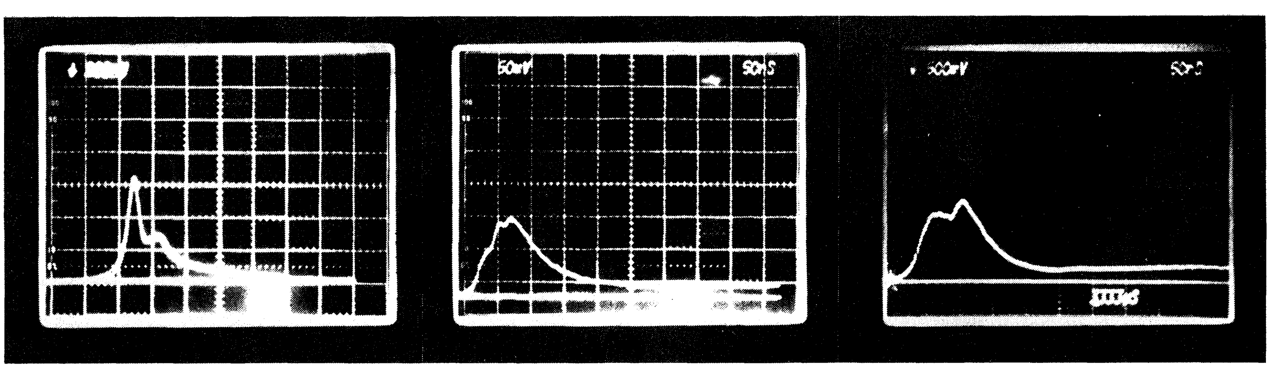

FIG. 5. Oscilloscope traces showing optical bistability. Cf. Figs. 4(c), 4(d), and 4(e).

ond order.

These results establish molecular gases pumped by CO_2 lasers as extremely promising media for the demonstration and investigation of chaos in all-optical systems. As well as a vast range of laser-molecule coincidences, there is an additional flexibility in that pressure, of both absorber and buffer, can be used to control response times. This flexibility should lead to operation with pulses long compared to t_R—even cw operation—which would make possible experimental verification of the predictions of Moloney, Hopf, and Gibbs[15] regarding the routes to chaos in systems of this type.

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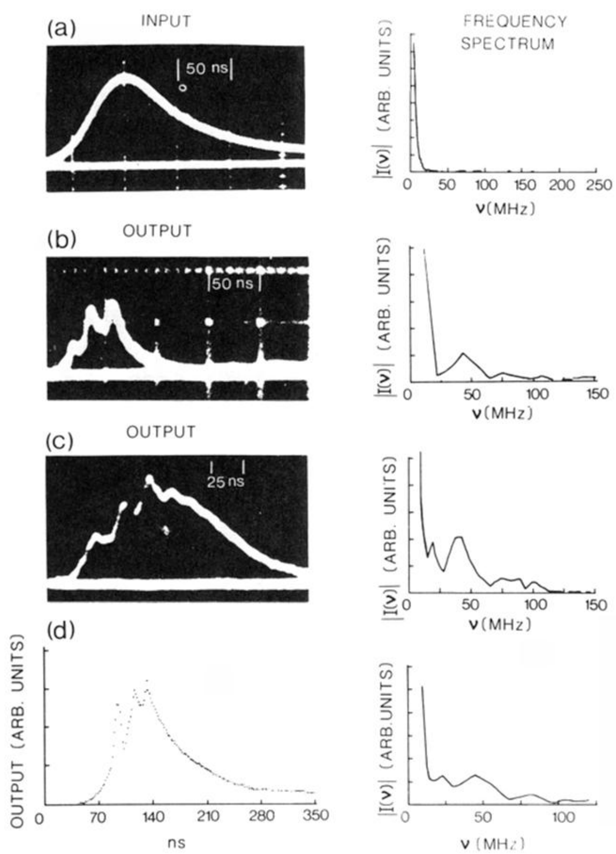

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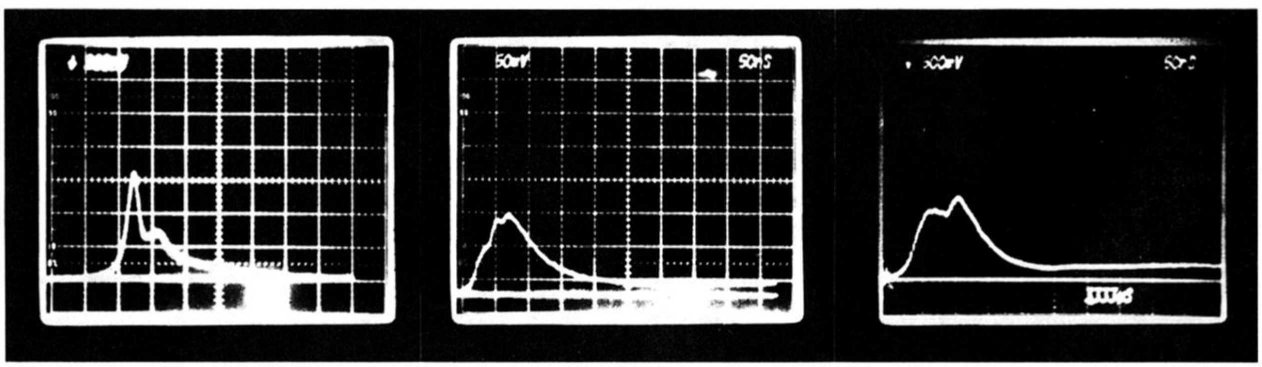

FIG. 5. Oscilloscope traces showing optical bistability. Cf. Figs. 4(c), 4(d), and 4(e).