Observation of optical hysteresis in an all-optical passive ring cavity containing molecular gas

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Dispersive optical hysteresis in an all optical passive unidirectional ring cavity containing a molecular gas has been observed. Ammonia is used as the nonlinear medium off resonantly excited on the aR (1,1) transition using temporally smooth 100-ns pulses from a transversely, excited, atmospheric pressure CO_2 laser. Results are in excellent agreement with the theory of Ikeda recently generalized by the authors to describe period doubling in this system.

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Nonlinear optical resonators have attracted great interest in recent years, with potential applications of optical bistability (OB) as a major motivation.[1] Semiconductors are claimed to have most device potential,[2] while atomic vapor,[3] hybrid,[4] and liquid crystal[5] systems have also attracted experimental and theoretical interest. The main object of this letter is to show that molecular gases have great, but largely unexplored, potential as devices in the OB field.

The particular system we describe here is an ammonia cell contained in a ring resonator, pumped by a single-longitudinal-mode, transversely, excited, atmospheric pressure (TEA) CO_2 laser, which typifies such systems. First, we have a near-resonant allowed transition ($\nu_{CO_2}-\nu_{NH_3}$ ~1 GHz), well removed (~5 cm^{-1}) from competing transitions, giving a clean, near two-level nonlinear medium with relatively simple dynamics. In particular, the transitions are homogeneously broadened at pressures of interest. Many other gases have near-resonant transitions with the CO_2 laser providing a range of frequency offsets well documented from photochemistry studies.[6] Second, pressure of the gas (and possibly buffer gas) can be used to control absorbance and response time. Response time control makes gases ideal media for investigation of instabilities and chaos in OB systems,[7] and we have recently reported[8] observation of period doubling (the first step to chaos) in our present system. A third, practical, advantage is our ability to operate at room temperature. Against these positive features, we must acknowledge that we operate with high power, relatively short TEA CO_2 laser pulses.

In this letter we report the first observation of dispersive optical hysteresis (OH) in a ring cavity containing a molecular gas. (We refer to OH rather than OB because, in our present system, the pump pulse length is not quite sufficient for switching transients to die out.)

There are many near coincidences between vibrational-rotational transitions of NH_3 and CO_2 laser lines[9,10]: for the present work we selected the aR (1,1) transition, which lies 1.23 GHz[9] below the 10R (14) CO_2 laser line.

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 (FWHM) ~100 ns [Fig. 2(a)] and peak power ~1 MW. The laser pulses were coupled, using a single-surface Ge flat ($R = 36\%$), into a 3.5-m three-element ring cavity, closed by 100% gold mirrors. The intracavity gas cell was one meter in length and terminated with Brewster angled KBr windows. The cavity geometry ensured unidirectional propogation with no feedback to the TEA CO_2 laser. The input and cavity signals were sampled by KBr beam splitters, and monitored by photon drag detectors and a Tektronix 7104 oscilloscope: total response time $\lesssim 1$ ns. For NH_3 pressures ~9–15 Torr, significant self-focusing was observed in single-pass experiments, confirming a nonlinear refractive index contribution ($n_2 \sim 2\times 10^{-9}$ esu) substantial enough for dispersive optical bistability. Closing the ring cause huge distortions of the pulse shapes (sampled after the NH_3 cell).

The oscilloscope traces in Fig. 2 are representative examples of the cavity signal showing strong pulse distortion indicative of optical hysteresis. We note that because our cavity was free standing, its length inevitably drifted from shot to shot, enabling us to sample the dependence of pulse shape on cavity mistuning θ. For the purpose of analysis independent absorption measurements were taken over an extended pressure range using short cell lengths of 10–20 cm, thereby minimizing self-focusing and ensuring essentially constant beam cross section throughout the cell length. Typical input and output pulse shapes are shown in Fig. 3. (Similar pulse shapes were obtained in the 1-m gas cell with the ring cavity blocked to prevent feedback).

The optically pumped ammonia is not quite a two-level system since allowance must be made for collisional population transfer within the rotational manifolds as described in detail elsewhere.[11]

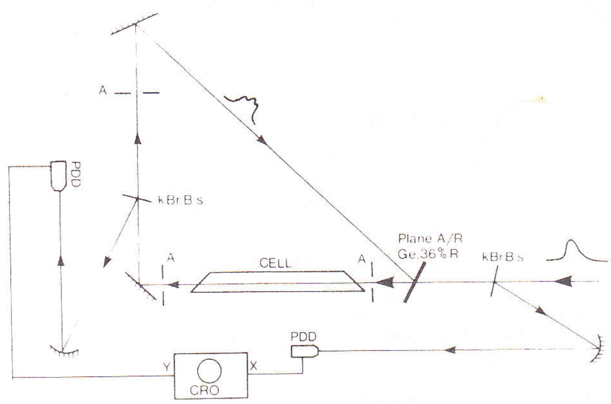

FIG. 1. Schematic diagram of ring-cavity system. BS: beam splitter, PDD: photon drag detector, A/R: antireflection coated.

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