Demonstration of Self-Pulsing Instability and Transitions to Chaos in Single-Mode and Multimode Homogeneously Broadened Raman Laser

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Single-mode and multimode self-pulsing instability leading to chaos is reported in the 12.8-μm emission from a homogeneously broadened NH_3 laser, near resonantly pumped by CO_2 laser radiation. Occurrence of instabilities on two independent emitting transitions over a wide range of operating conditions, including those for optimum lasing, indicate that this behavior is general for near resonantly enhanced two-photon Raman systems of this type.

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Here we report the first experimental observation of instabilities leading to chaos in a single-mode homogeneously broadened Raman laser system. These effects have been obtained on two independent emitting transitions over a wide range of operating conditions, including those for optimum lasing, which suggests that this behavior is indeed general for this broad class of near resonantly enhanced two-photon lasers. The route to chaos in this system is characterized by period doubling. Large variations in the basic pulsation period are found with changes in cavity fine tuning and the amplitude of these pulsations is itself modulated in some cases.

The laser, which uses ammonia gas as the active medium, selected on the basis of well documented discrete spectroscopic features, efficient lasing action, and previous demonstration of passive instabilities,[1] is optically pumped by pulsed CO_2 laser radiation. The pump pulse is steady for more than sixty pulsation periods of the instability, which thus establishes the steady-state nature of these effects. We concentrate on the $aP(8,0)$ lasing transition at 812 cm^{-1} optically pumped on the $aR(6,0)$ transition 1.3 GHz below line center by the $9R(16)$ CO_2 lasing emission at 1076 cm^{-1}. This laser transition has been clearly identified as Raman in origin[2] for NH_3 pressures ~ 1–20 torr and pump intensity of ~ 0.6 MW/cm^2. The pressure-broadened bandwidths of the pump and lasing transitions are 17.76 and 17.049 MHz/torr, respectively,[3] which, for our typical operating pressure of ~ 8 torr, considerably exceed the Doppler bandwidth (74 MHz), which nevertheless is essentially irrelevant because only one velocity group of the molecules is pumped.

A transversely excited atmospheric CO_2 laser was

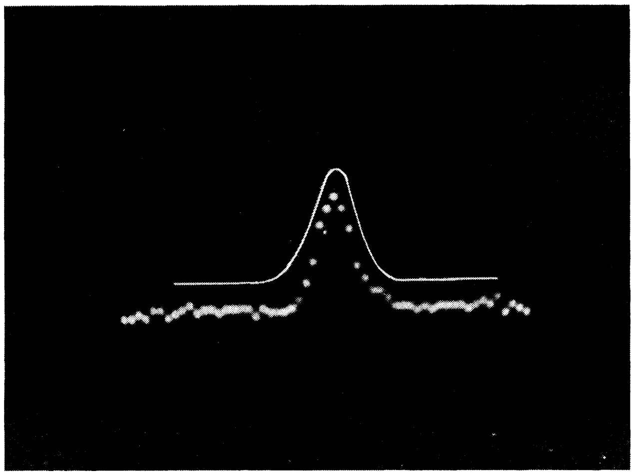

FIG. 1. Spatial intensity distribution of NH_3 emission taken with a pyroelectric array detector. A theoretical Gaussian profile is shown for comparison.

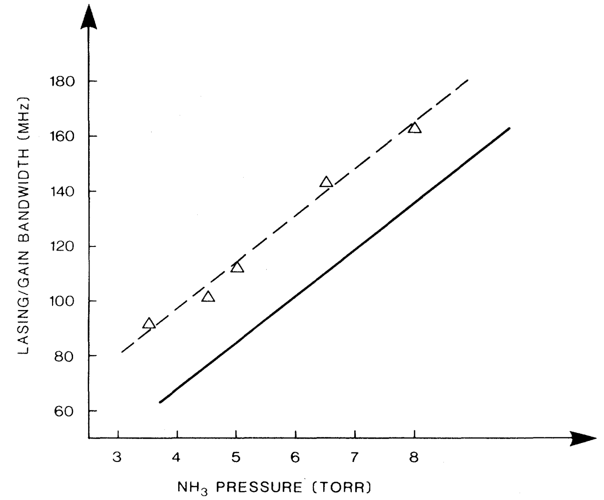

FIG. 2. Lasing and gain bandwidth of NH_3 emission as a function of pressure: dashed line, experimental data for lasing bandwidth determined from the PZT tuning range over which lasing occurred; solid line, prediction for gain bandwidth based on a FWHM value of 17.049 MHz/torr (Ref. 3).

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used as the pump source and operated on a single transverse and axial mode to generate temporally smooth long pulses (2-μsec FWHM with 250-kW peak power). Both the pump and the NH_3 signals were recorded on a fast detector and displayed on a Tektronix 7104 oscilloscope with response time ~ 1 nsec. The optically pumped Fabry-Perot cavity of length ~ 25 cm was provided with a piezoelectric tuning (PZT) facility and contained a KBr-Brewster-terminated NH_3 cell of length 22 cm. Details of the cavity optics are provided in the appropriate figure captions. Optical feedback to the CO_2 laser cavity was eliminated by slight tilting of the Fabry-Perot cavity with respect to the pump direction.

Transverse scans of the NH_3 emission taken by a pyroelectric array detector confirmed an essentially Gaussian spatial profile, which indicates operation on the lowest-order longitudinal mode. A typical recording is shown in Fig. 1 along with a theoretical Gaussian line shape for comparison. The form of the intensity distribution was found to be invariant to cavity length tuning; this insensitivity is attributed to the approximate mode-matched conditions of the pump and the TEM_{00} mode of our NH_3 laser cavity. The features of

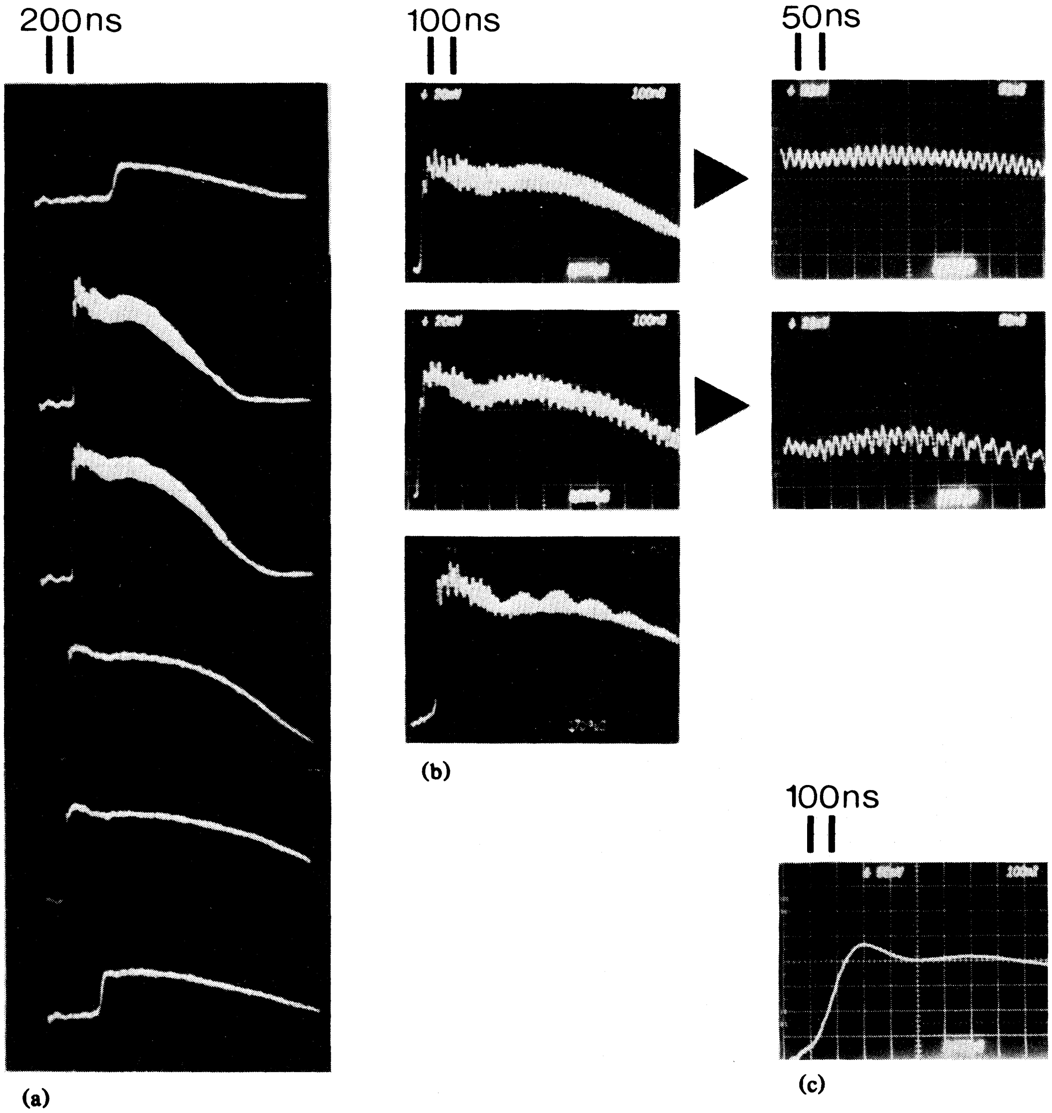

FIG. 3. (a) Cavity tuning scan across the region ($\sim \frac{1}{4}$ FSR) over which NH_3 lasing is obtained. (b) Examples of single-mode instability: long-periodic modulation (top trace); period doubling (middle trace); high-period chaos (bottom trace). The corresponding time-expanded traces are shown with an arrow. (c) Input pump pulse (cavity length 25 cm; mirror reflectivities 100% and 64% at 12.8 μm; NH_3 pressure ~ 8 torr).

the instabilities were maintained across the beam cross section. In subsequent data the whole beam signal was therefore monitored.

For a pump intensity of ~ 500 $\mathrm{kW/cm^2}$ lasing was obtained up to a pressure of 11 torr, which corresponds to a pressure-broadened gain bandwidth of ~ 187 MHz, substantially smaller than the free spectral range (FSR) of the laser cavity, viz., 600 MHz, thus ensuring a single-mode condition. This was further established from a determination of the cavity tuning range as a function of pressure over which lasing was obtained. The data of Fig. 2 are consistent with the predicted dependence of gain bandwidth on pressure shown as a solid line, based on a value of 17.049 MHz/torr. The displacement in the experimental data shows lasing to occur over a range somewhat greater than the FWHM of the gain bandwidth, which thus indicates a gain that is a little over double the lasing threshold.

Chaotic and periodic pulsation behavior in the $\mathrm{NH_3}$ emission was sensitive to cavity length tuning and occurred over $\mathrm{NH_3}$ pressures of 5–9 torr, smaller than the total range (3–11 torr) for lasing emission, with the most pronounced effects occurring at a pressure ~ 8 torr. A typical PZT scan (over $\frac{1}{4}$ FSR) at this pressure is shown in Fig. 3(a). Asymmetrical occurrence of these effects with respect to cavity tuning is similar to the observation of Weiss and Klische[4] for a far-infrared laser.

Within the narrow tuning range over which instability prevailed, two fundamental pulsation periods were found to occur for different PZT settings: one at ~ 3.8-nsec period and the other of relatively long period ~ 18 nsec. (The wave form was always found to be triangular.) These are shown in the time-expanded traces of Figs. 3 and 4. It is the slow periodic modulation [Fig. 3(b), top trace] which exhibited distinct period doubling [Fig. 3(b), middle trace] with very fine cavity length tuning before going into high-period chaos[5] [Fig. 3(b), lower trace].

In contrast, the high-frequency oscillations (Fig. 4) exhibited distinctly different behavior. Figure 4(a) shows a kind of intermittency[6] where there is a direct transition from an orderly state, here initial low-frequency modulation, into metastable chaos,[7] viz., a chaotic burst followed by an abrupt transition to a steady output. The beating effects seen in Fig. 4(b) can be identified with heavy breathing,[8] while Figs. 4(c) and 4(d) respectively typify examples of weak and fully developed chaos.[5]

The 3.8-nsec period of the high-frequency pulsations [e.g., Fig. 4(b)] is to be compared to the cavity round-trip time of 1.6 nsec. Straightforward mode-pulling considerations show that for a homogeneous gain bandwidth of 136 MHz (8 torr) this is consistent with intermode beating if the cavity linewidth is ~ 170 MHz (we recall here that transverse effects are not important for our observation). Calculation of the cavity

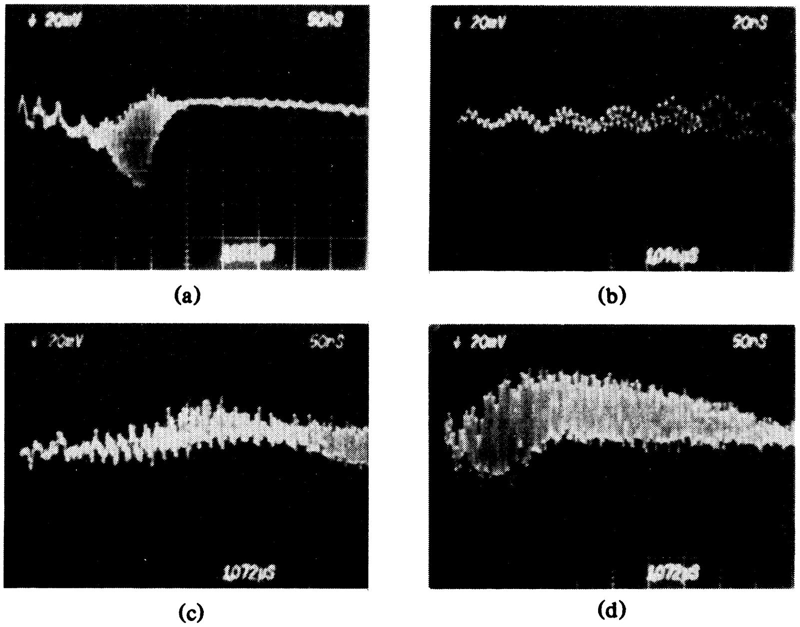

FIG. 4. Representative examples of high-frequency instabilities in the $\mathrm{NH_3}$ emission on cavity fine tuning. (a) Intermittent metastable chaos; (b) heavy breathing; (c) weak chaos; (d) fully developed chaos (cavity parameters as for Fig. 3).

linewidth, accounting for the NH_3 absorption and losses through the windows, indeed yields a value close to this. This further implies that we are observing bad cavity instability phenomena which are finally killed at the higher pressures ($\geqslant 10$ torr) where homogeneous broadening exceeds the cavity linewidth. Returning to Fig. 3, the oscillation period there is nearly 18 nsec with no evidence of high-frequency pulsations. The pulsations appear to be steady while the pumping is uniform, and this therefore suggests a clear case of single-mode instabilities at relatively low excitation above threshold (see Fig. 2).

The generality of our observation has been established from investigations of another NH_3 lasing transition pumped on the $aR(5,K)$ transitions 135 MHz off line center, notably in much closer resonance than that for the $aR(6,0)$ transition. At the only pump intensity used (~ 10 MW/cm^2), fully developed chaotic emission prevailed over all lasing conditions and this suggests that the onset of instabilities will occur at considerably reduced intensities. Indeed, the operation of this emission under cw conditions[9] provides exciting prospects for future investigation.

Test experiments, conducted with a dispersive Fabry-Perot system which could sustain only NH_3 emission, also gave laser instabilities, confirming that these effects arise from the active optical cavity alone.

Finally, we note the recent report of instabilities and chaos in a single-mode far-infrared (81.5 μm) optically pumped NH_3 ring laser.[10] Exact resonant excitation of the medium at low pump intensities ensures that lasing arises from population inversion rather than a Raman process. As the authors suggest, the instability phenomena that they observe may therefore be of the Lorentz type.

In conclusion, we have demonstrated huge pulsating instabilities with identifiable routes to chaos in the emission from a mid-infrared single-mode homogeneously broadened two-photon Raman laser at an excitation level much less than that considered necessary for two-level single-mode laser systems.[11] We note that these phenomena, investigated here for two commonly studied laser transitions in ammonia, are obtained over a wide range of operating conditions representative of those common to conventional laser systems of this class. Although quantitative understanding of two-photon Raman lasers, extensively used for mid-infrared to far-infrared generation, is in a somewhat embryonic state, our observations have identified them as specially suitable systems for the manifestation of instability phenomenon. We therefore hope that our results will stimulate further theoretical interest towards elucidating the dynamic behavior of these systems in regard to such effects.

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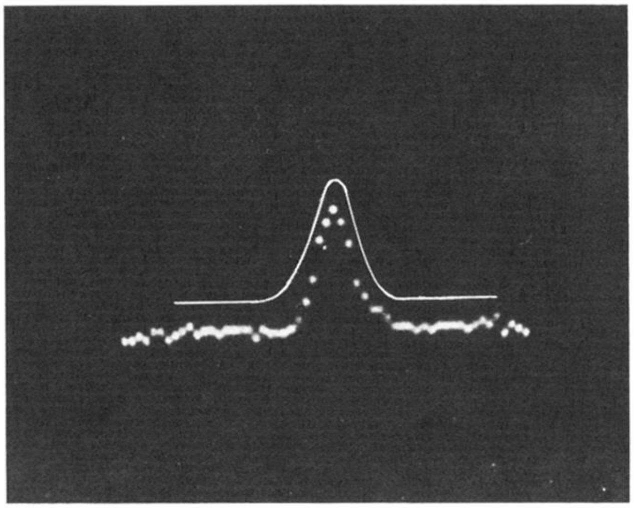

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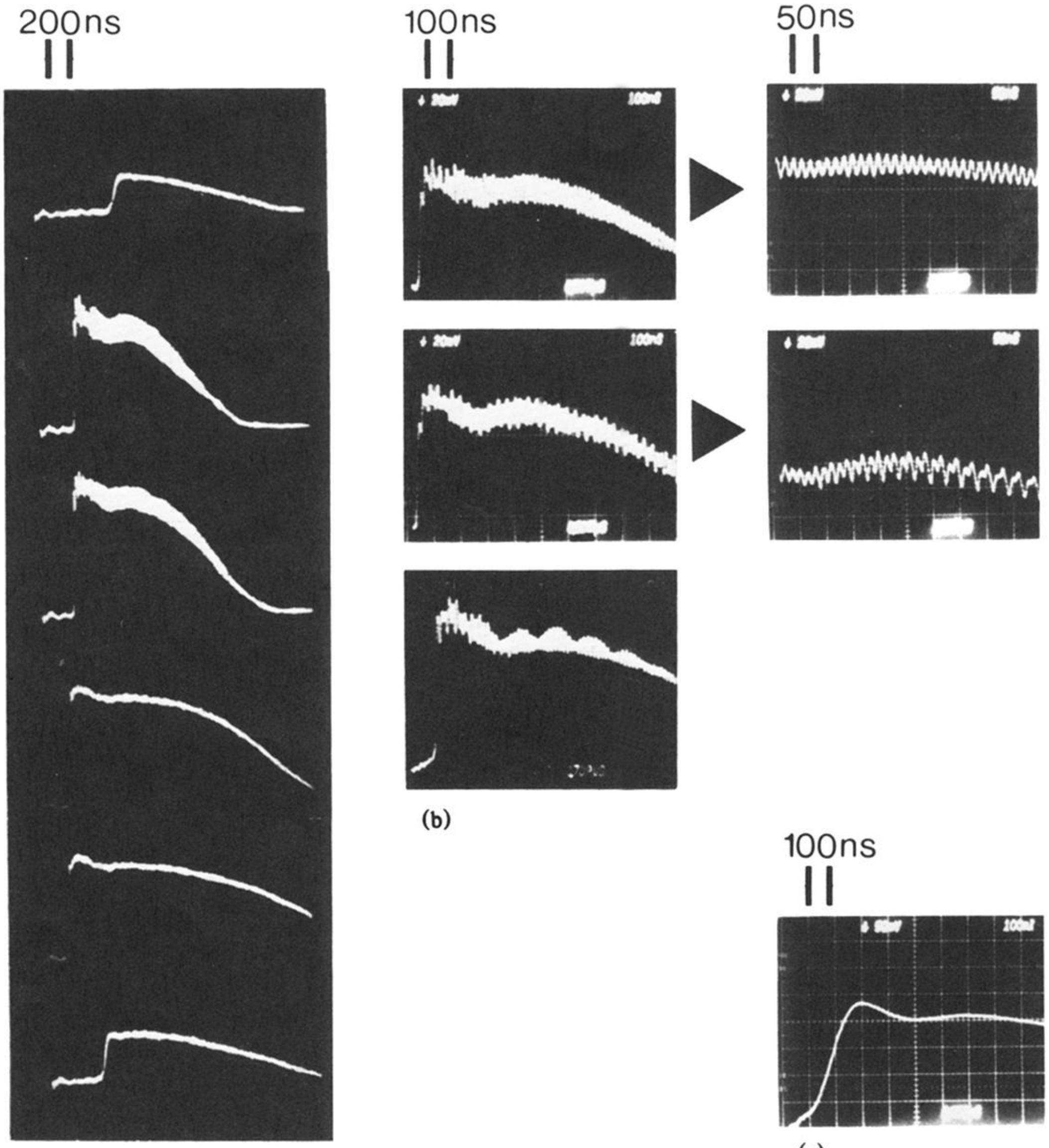

(a)

(b)

(c)

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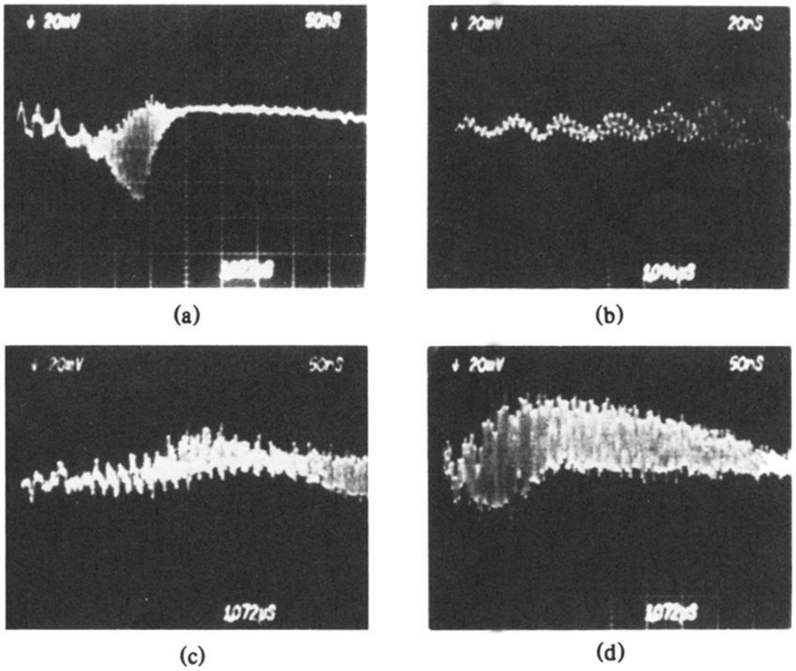

(a) (b)

(c) (d)

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