TWO-PHOTON PUMPING OF A FOUR-LEVEL SYSTEM IN AMMONIA TO OBTAIN 12.16 μm RADIATION FOR ISOTOPE SEPARATION*

John W. Leap, Kyung J. Kim, Edward G. Malk, and Paul D. Coleman
Electro-Physics Laboratory
Department of Electrical Engineering
University of Illinois
Urbana, Illinois 61801

ABSTRACT

Radiation at 12.16 μm (822.65 cm^{-1}) has been obtained from $^{14}NH_3$ using the $2\nu_2a(53)$ to $\nu_2s(63)$ transition with the upper level pumped from Ga(33) via $\nu_2s(43)$ by two CO_2 TEA lasers. In an unoptimized cavity, efficiency was 1%: 0.5 mJ of 12.16 mJ out with 50 mJ of input (25% $P_9(8)$ $^{13}CO_2$ and 75% $P_9(24)$ $^{12}CO_2$). This particular wavelength is relevant to LIS of uranium.

Introduction

Molecular optical pumping with CO_2 lasers is a prolific source of new infrared lasers. Hundreds of discrete wavelengths have been produced, but seldom to meet a specific design goal. Our successfully obtained objective was to efficiently produce a particular frequency (823 ±1 cm^{-1}) which is applicable to LIS of uranium. This frequency (822.65 cm^{-1}) was generated with one-photon pumping and even more successfully with two-photon pumping of $\nu_2a(53)$ in $^{14}NH_3$.

For a given desired output frequency, the optically pumped molecule must have an allowed transition at this frequency. For direct optical pumping, versus collisional or cascade pumping, the transition upper level must be shared with a transition which is nearly resonant with a CO_2 laser frequency as is shown in Figs. 1 and 2. Detailed spectroscopic data such as in Ref. 1, is essential for planning specific output experiments.

The $\nu_2s(43) \rightarrow 2\nu_2a(53)$ is a "hot-band" match with $P_9(24)$ $^{12}CO_2$ and Gullberg et al.[2] have observed lasing (35 and 65 μm) from $2\nu_2a(53)$ using a single $P_9(24)$ pump, but did not report 26.4 μm observed for the first time in one of our experiments. With an additional 26.4 μm loss mechanism present, the desired 12.16 μm (822.65 cm^{-1}) was observed.

More desirable pump threshold, efficiency, and output energy were observed with two-photon pumping shown in Fig. 2. Both CO_2 laser pump photons are nearly resonant (pump-absorption frequency mismatch < 0.01 cm^{-1}) and a two-step pumping process is likely. Two-step pumping actually populates the intermediate level $\nu_2s(43)$, whereas one-step two-photon absorption does not. Two-step pumping

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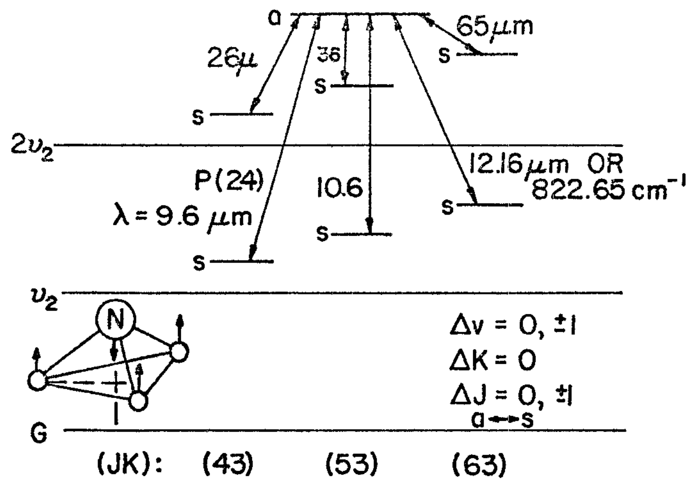

Fig. 1. Allowed transitions from $2\nu_2a(53)$ to lower states.

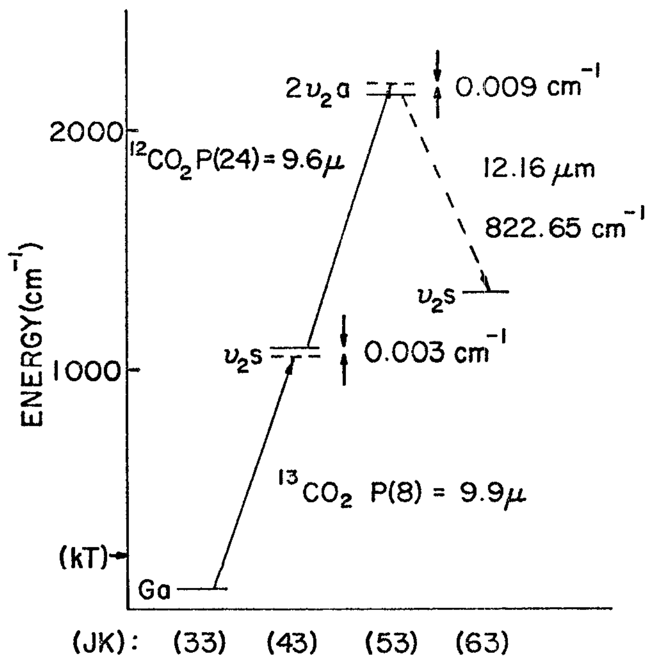

Fig. 2. Two-photon pump frequency mismatches.

should be easy to achieve with such small mismatches and any companion one-step pumping would be a bonus. Either mechanism uses the strongly populated starting state Ga(33). At room temperature, the two most heavily populated states are Ga(33) and Gs(33).

Jacobs et al.[3] sought and apparently effected one-step, two-photon pumping of $2\nu_2a(54)$ in NH_3 to produce ten weak ($\lesssim 1$ μJ emissions from 6 to 35 μm. Their pump frequencies were individually off-resonant (mismatch > 0.1 cm^{-1}), but cummulatively had a mismatch of only 0.01 cm^{-1}. Those same authors[4] similarly produced 9.75 μm emission from $^{12}CH_3F$.

In search of an 823 ±1 cm^{-1} laser, the following was done:

1) identified a useable transition in $^{14}NH_3$, $2\nu_2a(53) \rightarrow \nu_2s(63) = 822.65$ cm^{-1},

2) with one-photon pumping,

a) observed strongly dominant (even superfluorescent) competitive 26.4 μm emission,

b) suppressed or discouraged this competing emission sufficiently to observe the desired 822.65 cm^{-1},

3) with two-photon pumping,

a) generated 822.65 cm^{-1} in an unoptimized cavity with an efficiency of 1%, two orders of magnitude better than with one-photon pumping,

b) briefly studied pump pulse threshold and synchronization.

Apparatus

The main experimental arrangement is shown in Fig. 3. The moderately short (0.7m) cell length and the intracavity ZnSe Brewster window beam splitter, together discouraged emission of all wavelengths > 20 μm. All mirrors shown were gold-coated glass (broadband). Both CO_2 TEA lasers were laboratory-built and driven lightly (∿ 20 joules input) using hydrogen thyratrons (7666/KU73) for capacitor switching. These have a low-jitter, high-pulse repetition frequency capability which was used to synchronize or relatively delay pump pulses. The $^{12}CO_2$ laser used a flowing gas mixture and had a $^{12}CO_2$ CW gain cell in its optical cavity to allow single longitudinal mode operation. The $^{13}CO_2$ laser used a refillable sealed static gas mixture and was operated modelocked. Typical pulse waveforms (modelock structure omitted) are shown in Fig. 3. Output energies were typically 60-200 mJ and 10-30 mJ for P(24) and P(8), respectively.

Results

One-photon pumping in a simple (no ZnSe filter window) 1 meter hole-coupled cavity or in a single-pass 6 mm I.D. Pyrex waveguide did not generate 12.16 μm because the competing 26.4 μm (shown in Fig. 1) was dominant. Changing to the cell shown in Fig. 3,

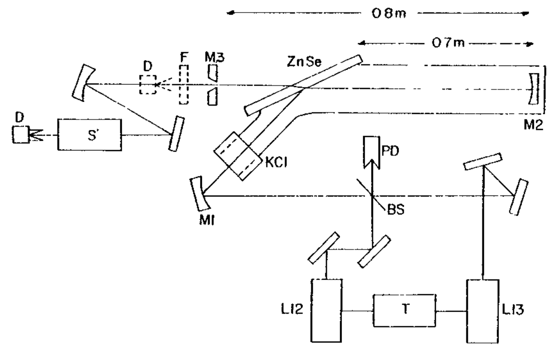

Fig. 3. Experimental arrangement for two-photon pumping with typical pump waveforms. A: ammonia cell; BS: beamsplitter; D: detector (pyroelectric or HgCdTe); F: filter (passes $\lambda \geq 12$ μm); L12: $^{12}CO_2$ laser; L13: $^{13}CO_2$ laser; M1: mirror (R=0.7m); L13: mirror (R=4m); M3: flat mirror with 0.5 mm hole; PD: photon drag detector; T: trigger.

yielded the desired 12.16 μm emission. The P(24) pump threshold for 12.16 μm lasing with 8 torr of NH_3 was ∿ 50 mJ singlemode or ∿ 100 mJ modelocked. The best 12.16 μm output was ∿ 50 μJ.

Compared to single-photon pumping, two-photon pumping (shown in Fig. 2) had a lower pump threshold: ∿ 1 mJ P(8) modelocked plus ∿ 3 mJ P(24) single mode, versus ∿ 50 mJ P(24) single mode, and had a lower NH_3 optimum pressure: ∿ 1 torr versus ∿ 8 torr.

The two-photon experiment was very successful in generating 12.16 μm, plus smaller amounts of 10.6 μm and 12.6 μm radiation in relative energy ratios of ∿ 10:1:<1, depending on experimental conditions. Signal waveforms are shown in Fig. 4. The 12.6 μm lasing is a desirable cascade process which depletes $\nu_2s(63)$. Only the 10.6 μm emission is detrimental. The measured 12.16 μm output energy was 0.5 mJ for pump pulse energies of 25 mJ P(8) and 70 mJ P(24). Of this 95 mJ total, only half was usefully injected along the NH_3 cavity axis, for an unoptimized conversion efficiency $\eta \approx 1\%$.

$$\eta = \frac{\text{output energy}}{\text{input energy}} = \frac{0.5\text{mJ}}{(0.5)(95\text{mJ})} \approx 1\%$$

The Manley-Rowe theoretical efficiency for this two-photon pumping process is ∿ 40%.

An indication of two-step pumping is the appearance of 12.0 μm lasing from the intermediate level $\nu_2s(43)$ when only the P(8) pump

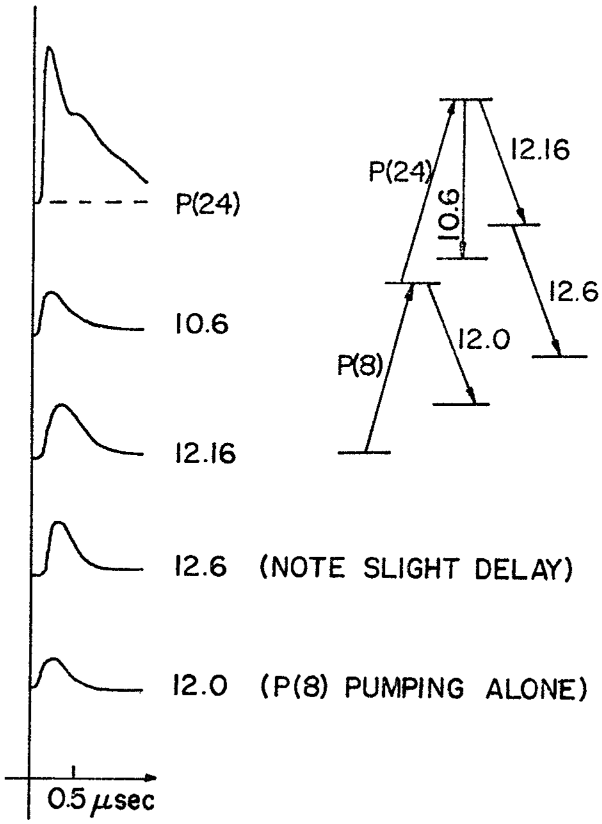

Fig. 4. Waveforms of lasing signals from 2 torr NH_3, via monochrometer to HgCdTe detector. P(24) preceded P(8) by 0.1 μsec.

is present. Leite et al.[4] have summarized and confirmed various relaxation times associated with a different state $\nu_2 s(87)$. By analogy, a population decay time $T_1 \sim 0.1$ μsec (at 1 torr) is expected for $\nu_2 s(43)$. Such a T_1 with two-step pumping would not be inconsistent with the observed effect of relative P(8)-P(24) pulse delay on 12.16 μm output, shown in Fig. 5.

Since strong (> 10^5 watts/cm^2) nearly-resonant fields are present, a simple model, such as two-step pumping is not expected to be more than a partial description. A four-level, three-wave density matrix steady-state computer model, including higher-order interactions, is being developed, but is not available at this time.

Conclusion

Although better conditions, such as optimum percentage output coupling with a frequency selective, full-aperture, dielectric coated mirror, could improve efficiency, two-photon pumping of $^{14}NH_3$ with CO_2 lasers has efficiently (1%) produced 0.5 mJ, 0.5 μsec pulses of radiation at a premium frequency (822.65 cm^{-1}) desirable for uranium isotope separation.

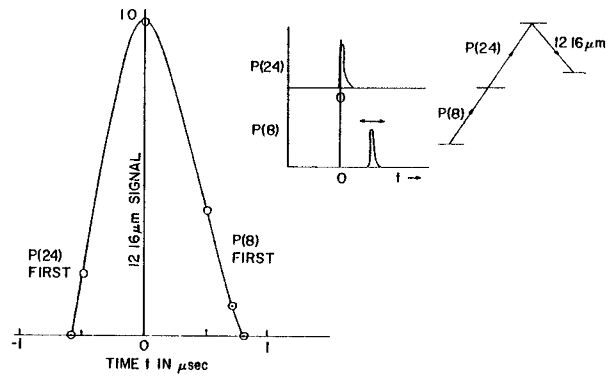

Fig. 5. Effect on signal of relative time delays of pumps. Ammonia pressure 1 torr, pumps modelocked with reduced nitrogen tail.

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