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TITLE: OPTICAL PUMPING OF THE $\nu_3 + \nu_4$ BAND OF THE CF_4 MOLECULE

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Optical Pumping of the $\nu_3 + \nu_4$ Band of the CF_4 Molecule

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Abstract

The possible development of a room-temperature, 16 μm CF_4 laser optically pumped by 5 μm radiation from a frequency-doubled CO_2 laser has been investigated. This laser would utilize pumping of the $\nu_3 + \nu_4$ band of CF_4 at 1916 cm^{-1} with subsequent lasing to ν_3 levels at 1283 cm^{-1} where there is essentially no thermally excited population. A spectrophone instrument was used to measure absorption cross sections of CF_4 at the second harmonic frequencies of 22 of the 10 μm CO_2 laser transitions. Lasing experiments to detect transitions at 16 μm are described.

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The CF_4 molecule, when optically pumped by 9 μm CO_2 laser transitions, has been one of the most powerful and spectrally rich of the mid-infrared lasers.[1] The strongest lasing transition reported thus far occurs at 615.1 cm^{-1} when CF_4 is pumped by the 9R(18) CO_2 transition. Other frequencies have been observed ranging from 600 cm^{-1} to 655 cm^{-1} with the CO_2-pump transitions varying from 9P(14) to 9R(24).[2] In fact, a single CO_2 pump transition may result in several CF_4 lasing frequencies if the CO_2 oscillator is tuned across its full gain bandwidth of several GHz.

This multiplicity of CF_4 transitions results from absorption between the molecular ground state and the $\nu_2 + \nu_4$ combination band at 1061 cm^{-1} with subsequent lasing to the ν_2 manifold at 435 cm^{-1}. Figure 1 depicts the pertinent vibrational levels of CF_4 with the above-described process illustrated on the left side of the figure. The richness of the CF_4 laser results from pumping different levels of the $\nu_2 + \nu_4$ rotational-vibrational manifold.

The lower lasing levels of this process are ν_2 rotational levels with energies in the neighborhood of 435 cm^{-1}. Thus, the gas must be cooled to prevent thermal excitation from spoiling any population inversion that could be produced by pumping $\nu_2 + \nu_4$. Typically, the CF_4 cell is cooled to about 130°K by placing it over a liquid-nitrogen reservoir. Generally, it has been difficut to achieve any detectable lasing in room-temperature CF_4 on the $(\nu_2 + \nu_4) \rightarrow \nu_2$ transition.

We have attempted to overcome this difficulty by pumping the $\nu_3 + \nu_4$ band at 1916 cm^{-1} with radiation derived by generating the second harmonic of various CO_2 transitions in the 10 μm band. In this case, lasing would take

place to the ν_3 levels around 1283 cm^{-1}. There is essentially no thermally excited population in this band at room temperature. This process is also illustrated in Figure 1. It is expected that an even richer group of 16 μm lasing transitions might result from pumping the spectroscopically more complex $\nu_3 + \nu_4$ manifold.

In addition to the possible laser transitions in the 16 μm range, lasing can take place to the ν_4 level at 633 cm^{-1} producing transitions in the 7.8 μm region. This is illustrated to the right of Figure 1.

(Figure 1) ~~4~~ 2)

To investigate the pumping mechanism we have utilized a spectrophone instrument to make measurements of the absorption cross sections of the molecule at the second harmonic frequencies of 22 of the 10 μm CO_2 laser transitions. The experimental arrangement for these measurements is illustrated in Figure 2. The 10 μm radiation from a single-transverse mode, grating-tuned, TEA CO_2 laser with about 800 millijoules output energy was doubled in frequency with a $CdGeAs_2$ crystal.[3] Typical energies of the 5 μm radiation ranged from 2 to 6 millijoules with pulse widths (FWHM) of approximately 150 nsec. A sapphire plate was then used to filter out the remaining 10 μm radiation and this reduced the 5 μm energy by about 50%.

The 5 μm radiation was passed through a 10-cm length spectrophone cell[4] having an electret microphone and NaCl Brewster windows of 2.5 cm diameter aperture. The remaining 5 μm radiation was monitored with a Gentec pyroelectric energy detector. Both the second-harmonic and the spectrophone-cell signals were displayed on a dual-beam storage scope. The CF_4 pressure was 10 torr and 10 torr of He was added to increase thermal conductivity. No signal was detected when 20 torr of He alone was used.

We calibrated our spectrophone response by comparing our acoustic-signal absorption measurements to that measured optically for the CO_2 9R(14) absorption in CF_4 as measured in reference 6. The CO_2 transitions used, their doubled frequencies, and the relative and absolute absorption coefficients are listed in Table I. The absorptions vary by a factor of 20 with the strongest absorptions occurring for the second-harmonic frequencies of the 10P(6) and 10P(20) transitions.

Table 1.....

Lasing experiments are under way utilizing a 2-meter optically-pumped CF_4 cell as illustrated in Figure 3. The CO_2 laser is a Lumonics 103, 1-joule, single-transverse mode TEA laser. After passing the resulting signal through a one-half meter Jarrell-Ash spectrometer, the lasing frequency is focused onto a HgCdTe detector.

Work, to date, has concentrated on pumping the CF_4 with doubled 10P(6), the 5 μm radiation with the strongest absorption. While lasing in the 7.8 μm region has been tentatively observed, further searching in the 16 μm region is still in progress. Since our pump absorption coefficients are only approximately 10 times lower than typical absorption coefficients reported for the $\nu_2 + \nu_4$ band,[5] it is hoped that room temperature lasing of CF_4 at 16 μm will soon be achieved.

References

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4. The spectrophone instrument was provided by Mel Buchwald, Los Alamos Scientific Laboratory, Los Alamos, NM.
5. J. J. Tiee and C. Wittig, Appl. Phys. Lett. 30, 420 (1977).

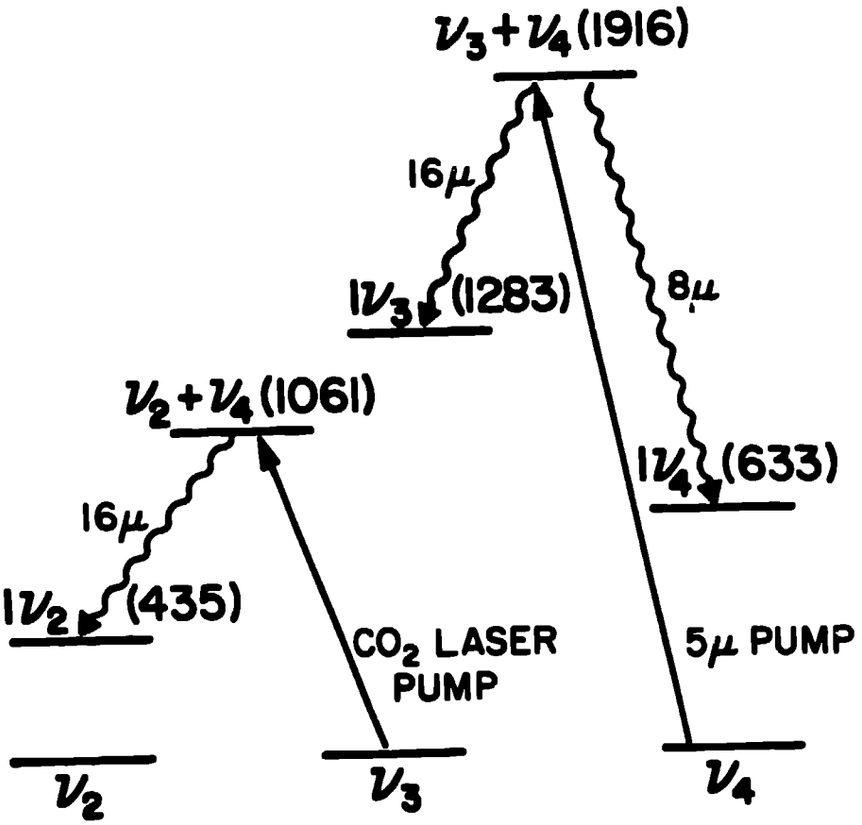

Fig. 1. CF_4 Energy Levels

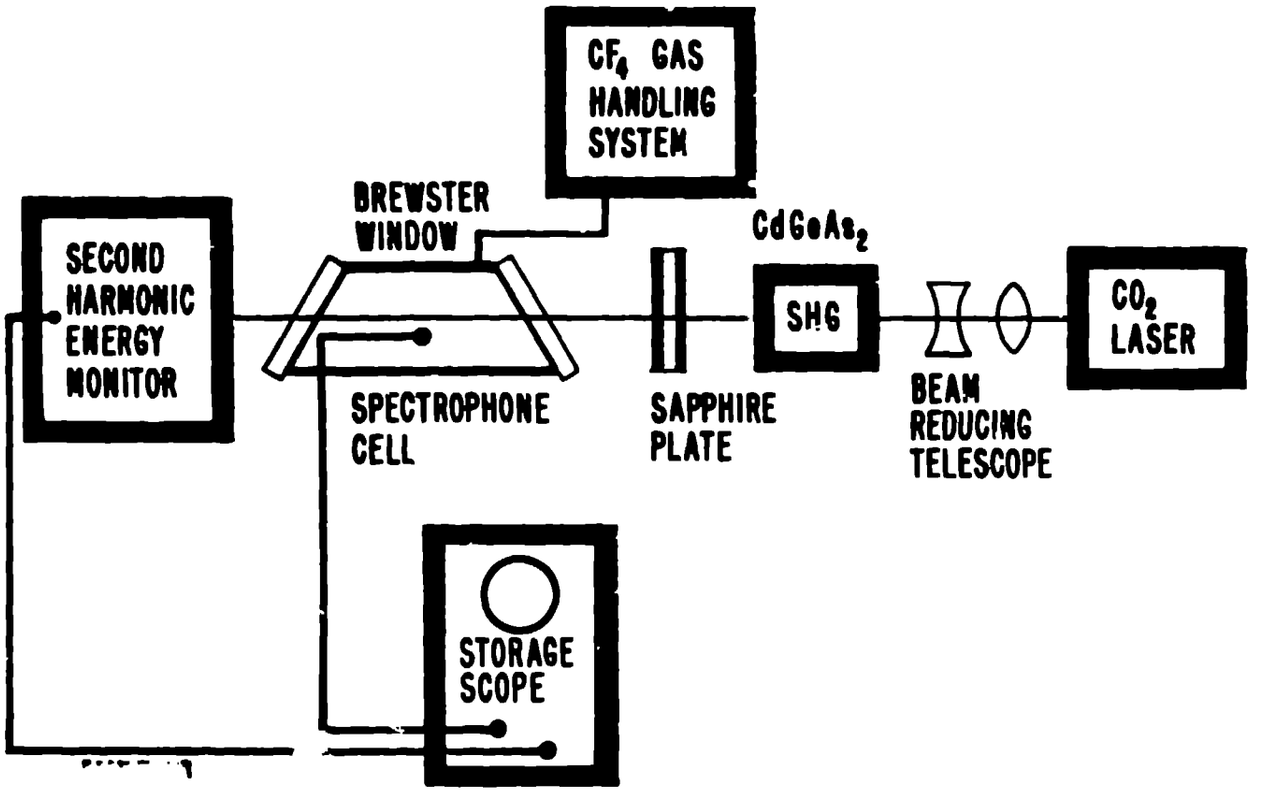

Fig. 2. Experimental Arrangement For Measurement of Absorption Cross-Section of $\nu_3 + \nu_4$ Band of CF_4

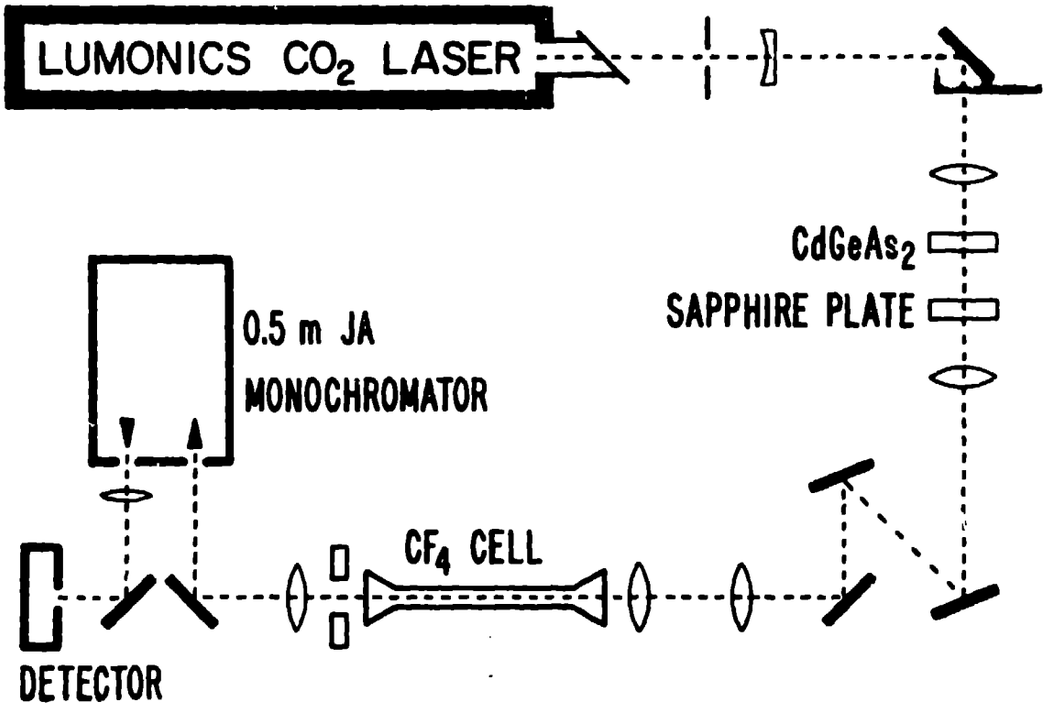

Fig. 3. $2_3 + 2_4$ Lasing Arrangement

Table I

CF_4 ABSORPTION COEFFICIENTS FOR $\nu_3 + \nu_4$

$^1_2$ LINE	DOUBLED FREQUENCY (cm^{-1})	RELATIVE ABSORPTION	ABSORPTION COEFFICIENTS (cm^{-1} $torr^{-1}$)
P(36)	1858.04	0.06	5.89×10^{-6}
P(34)	1862.00	0.18	1.76×10^{-5}
P(32)	1865.92	0.32	3.14×10^{-5}
P(30)	1869.79	0.10	9.8×10^{-6}
P(28)	1873.61	0.32	3.14×10^{-5}
P(26)	1877.38	0.40	3.93×10^{-5}
P(24)	1881.10	0.30	2.95×10^{-5}
P(22)	1884.77	0.92	9.03×10^{-5}
P(20)	1888.39	1.00	9.8×10^{-5}
P(18)	1891.96	0.80	7.86×10^{-5}
P(16)	1895.49	0.60	5.89×10^{-5}
P(14)	1898.96	0.75	7.37×10^{-5}
P(12)	1902.39	0.79	7.76×10^{-5}
P(10)	1905.76	0.68	6.68×10^{-5}
P(8)	1909.10	0.80	7.86×10^{-5}
P(6)	1912.37	1.2	1.18×10^{-4}
R(8)	1935.42	0.78	7.66×10^{-5}
R(10)	1938.28	0.60	5.89×10^{-5}
R(12)	1941.10	0.40	3.93×10^{-5}
R(14)	1943.86	0.18	1.76×10^{-5}
R(16)	1946.58	0.10	9.8×10^{-6}
R(18)	1949.25	0.08	7.86×10^{-6}