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# A TUNABLE FAR INFRARED LASER

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### Abstract

A continuously tunable far infrared (FIR) laser has been demonstrated; experimental results are presented. A high pressure (10-12atm) continuouslytunable CO<sub>2</sub> TE laser is used to pump Raman transitions in CH<sub>3</sub>F; the generation of continuously tunable radiation in the  $250\mu$ m- $300\mu$ m wavelength range is reported. Accurate frequency and bandwidth measurements have been made and the FIR bandwidth in superradiant emission is ~4-5GHz. Consequently, the generation of frequency tunable, subnanosecond pulses in the FIR appears feasible. The generation of tunable laser radiation from  $150-1000\mu$ m by stimulated Raman scattering should be possible using higher pump intensity and/or other gases.

#### Introduction

We present experimental results on a continuously tunable, high power far infrared (FIR) laser. Recently, there has been significant progress towards the goal of achieving tunable high power laser radiation in the FIR spectral region. Stimulated Raman scattering in the HF and HCl has been employed to generate tunable radiation in the 40-250µm spectral range.<sup>1,2,3</sup> Biron et al.<sup>4</sup> first demonstrated the feasibility of generating tunable FIR radiation by the stimulated Raman scattering (SRS) of CO<sub>2</sub> pump laser radiation in molecular gases. With the use of a highly focused pump beam and a dielectric waveguide for the FIR laser, FIR emission was observed on many Raman transitions in  $^{12}CH_{3}F$  and  $^{13}CH_{3}F$  with offsets of up to 30GHz from resonance.<sup>5</sup> Since that time, the theory describing these tunable Raman lasers has been derived,<sup>5,6,7</sup> and several experiments with continuously tunable CO<sub>2</sub> pump lasers have produced tunable FIR radiation.<sup>8,9,10,11,12</sup> We present experimental results on a  $^{12}CH_{3}F$ waveguide laser which is tunable from 250µm-300µm.

In this tunable  $CH_3F$  laser, the  $CO_2$  laser pumps molecules from the vibrational ground state (g,J), where J is the total angular momentum quantum number, to an excited state level  $(\upsilon_3, J^{\prime})$ . The pump transition can be a P, Q, or R branch transition, corresponding to  $J^{\prime} = J-1$ , J, J+1. The FIR transition occurs between rotational levels  $(J^{\prime} + J^{\prime}-1)$  within the excited vibrational state. The laser pumped emission process occurs via the coherent, Raman process. The Raman transition is denoted by the pump transition (P,Q, or R) and the initial J value.

## Experimental Results

The experimental configuration is shown in Fig. 1. The pump laser is a UVpreionized 10-12atm CO<sub>2</sub> TE laser. The main discharge is driven by a five stage Marx bank which produces 150kV, 36J pulses with a measured 10-90% risetime of llns, an internal inductance of 122nH and an output impedance of 11.5 $\Omega$ . The laser optical cavity consists of a 150%/mm diffractiion grating, a Germanium output coupler, and two telescopic beam expanders for the grating and output coupler. Laser output energy ranged from 40-120mJ, depending on Marx bank voltage and the emission frequency. For emission near the band centers of the 9 $\mu$ m R and 10 $\mu$ m R branches, the laser produces 80-100mJ reliably. For the 9 $\mu$ m P and 10 $\mu$ m P branches, only 40-80mJ near the line center frequencies is obtained. Continuously tunable output was obtained on the 9R and 10R branches, but on the 9P and 10P branches the gain was adequate only for tuning within  $\pm$ 5GHz about the line center frequency. The laser output pulse duration is typically ~100ns, and the bandwidth is ~4GHz. The high pressure CO<sub>2</sub> laser is described in detail elsewhere.<sup>6</sup>

The output of the pump laser is focused into a FIR waveguide laser through a NaCl window (W1). A fused quartz dielectric waveguide was used to lower the diffraction losses in the FIR and thereby reduce the threshold for stimulated Raman scattering. The  $CO_2$  beam is coupled into the  $EH_{11}$  mode of the dielectric waveguide by adjusting the pump beam radius at the entrance to the waveguide<sup>13,14</sup>. The FIR emission also propagates in the  $EH_{11}$  mode, as this is the lowest loss mode for this waveguide. In the experiments reported here, quartz tubes of diameter 7mm and 5mm and length 1.2m were used as the FIR waveguide. Even with optimum coupling between the free space pump beam and the  $EH_{11}$ waveguide mode, the pump beam transmission in an evacuated waveguide was limited

to about 70%. This limitation is due to a slight departure from straightness of the waveguide bore.<sup>15,16</sup> The FIR emission exits the FIR laser through a Teflon window.

Absorption of the pump beam by the FIR laser gas is monitored by a photoacoustic transducer (P.A.T.) mounted perpendicular to the optical axis near the entrance to the waveguide. For low pump powers, there is no AC Stark splitting; thus, when the acoustic signal is maximized, both the pump and emission fields are resonant. All frequency shifts resulting from the Raman tuning of the emission are then measured relative to those line center frequencies. Because the spectroscopy of  $CH_3F$  is known to high accuracy,<sup>17</sup> frequency shifts measured relative to line center can be accurately converted to absolute frequencies with the available spectroscopic data.

The measurement of the Raman tuning behavior of the CH<sub>3</sub>F waveguide laser was carried out in two steps. First, laser emission at a known frequency, such as that corresponding to line center emission, was obtained. Then the shift in FIR emission frequency was measured for a change in the pump laser frequency. In this manner, a set of pump and FIR frequency shifts from line center is obtained; this data yields tuning curves of  $v_s$  (FIR emission frequency) versus  $v_p$  (pump frequency). These turning curves can be compared directly with theory.

The measurement of frequency shifts for the pump and FIR beams was determined by several methods. Both the absolute FIR emission frequency and the frequency shifts in the FIR are determined by a scanning Fabry-Perot interferometer (II). This interferometer system consists of two wire grid inductive meshes, one of which is mounted on a motor-driven translation stage, a pyroelectric detector, and signal averaging electronics. The FIR interferometer was calibrated with the 496.1 $\mu$ m Q(12) emission line of CH<sub>3</sub>F.

A resonable estimate of the  $CO_2$  laser frequency could be made using the

grating angle, since frequency pulling effects were found to be small. A more accurate method for measuring the pump laser frequency shift has been recently incorporated into the experimental system; it involves the use of a piezoelectrically driven infrared Fabry-Perot interferometer (I2). This interferometer consists of two  $\lambda/100$  dielectric-coated Germanium mirrors with reflectivity 96%. The overall finesse of this instrument was measured with a CW CO<sub>2</sub> waveguide laser to be 42. Alignment of the Germanium optics proved difficult and limited the maximum finesse to this value. Nevertheless, this interferometer is adequate for measuring frequency shifts of 1GHz or more. The interferometer monitors the pump laser frequency by detecting a small portion of the pump beam which is reflected off the NaCl window (W1).

Using the high pressure  $CO_2$  pump laser and FIR waveguide laser, frequency tuning experiments in CH<sub>3</sub>F were performed. The FIR emission frequency was measured as a function of the pump laser frequency for P and R branch Raman transitions in CH<sub>3</sub>F.

The best demonstration of the generation of widely tunable FIR radiation from a CH<sub>3</sub>F Raman laser occurs for the R branch. For these transitions the available pump laser power is high, and the threshold for Raman emission is low.<sup>5,6</sup> Experimental results for R branch frequency tuning in CH<sub>3</sub>F are shown in Fig.2. FIR output power in the 1-10kW range was obtained; this corresponds to an efficiency of approximately 0.5%. In Fig.2, the FIR emission frequency is plotted as a function of the pump laser frequency for Raman emission on the R(J=19) to R(J=22) transitions. The experimental error is shown in the upper left; the pump frequency was determined from the CO<sub>2</sub> laser grating position. The theoretical tuning is shown (dashed lines), and the locations of the line center absorptions for each transition are indicated. The far infrared emission was tunable from  $33cm^{-1}$  to  $39cm^{-1}$  ( $256\mu$ m- $300\mu$ m). As predicted,<sup>5,6</sup> the

tuning is asymmetric about the absorption line center, with negative pump offsets favored. This is due to an interference between ground state and excited state Raman processes, both of which contribute to the gain at the FIR emission frequency. The discontinuity in Raman tuning occurs at a pump laser frequency just above the absorption line center.

Experimental results for Raman tuning on the P branch in CH<sub>2</sub>F have also been obtained; they are shown in Fig. 3. Because of the higher thresholds for P transitions at large J and the lower CO<sub>2</sub> laser output power available from our laser in this region, Raman tuning data was only obtained on the P(35) transition. The location of the P(35) line center absorption and the  $10.2\mu m$ R20 CO<sub>2</sub> laser transition are also indicated in Fig. 3. Data shown as circles are referenced to the CO2 laser line frequency; the pump laser frequency shifts were measured by the interferometer for this data. The data shown as squares are referenced to the P(35) absorption line center; the pump frequency shifts were calculated from the grating position for this data. Experimental error for the Stokes frequencies is ±1.5GHz in both cases. Errors for the pump frequencies are  $\pm 1.75$  GHz and  $\pm 3$  GHz for the circled and boxed data points. respectively. Raman tuning by  $\pm 4.5$ GHz was obtained about the line center, this range being limited by the available pump intensity. The data in Fig. 3 serve to demonstrate the generation of tunable FIR radiation on P transitions in CH3F. These transitions allow additional frequencies to be obtained beyond those available with R-branch tuning.

The presence of AC Stark shifts in a tunable FIR laser can result in a nonlinear tuning of the FIR emission frequency with the pump frequency.<sup>6</sup> Our experimental conditions precluded any determination of the relevance of the AC Stark shift to frequency tuning in this FIR laser. For the pump laser intensity of  $\stackrel{<}{\sim}$  6MW/cm<sup>2</sup>, the AC Stark shifts should be  $\stackrel{<}{\sim}$  1.4GHz. For the pump intensities,

pump bandwidth (4-5 GHz) and experimental error encountered in this investigation, the AC Stark shift, if present, is relatively small and difficult to measure. However, the effects of AC Stark shifts on frequency tuning curves may be important and measurable for high pump intensities or narrow-bandwidth, high resolution studies of tunable FIR Raman lasers.

The FIR laser bandwidth was measured as a function of the CO<sub>2</sub> laser pressure and is shown in Fig. 4. The bandwidth of the pump laser radiation increases with increasing laser gas pressure. For atmospheric pressure CO<sub>2</sub> lasers, both the pump and FIR laser bandwidths are typically  $\leq$  1GHz. However, for high pressure (≈10atm) operation of the pump laser, which is necessary to obtain frequency tunable emission, both the pump and FIR laser emission bandwidths were measured to be ≈4-5GHz. This represents the first determination of the bandwidth of a tunable FIR laser. These results indicate that the production of frequency tunable subnanosecond pulses in the FIR may be possible with similar laser pumped molecular lasers. Such a subnanosecond FIR laser, being both compact and relatively inexpensive, would compare favorably with FIR free electron lasers.<sup>18</sup> The existence of a simple, tunable high power, short pulse FIR source could benefit solid state physics research significantly.<sup>18</sup>

## Conclusions

The experimental results presented here not only demonstrate Raman tuning on R and P branch transitions, but they also demonstrate the feasibility of tunable far infrared generation by stimulated Raman scattering in polyatomic molecules. The straightforward extension of these principles to other FIR laser gases should result in the production of tunable laser radiation at other FIR frequencies. Alternatively, the availability of a higher power pump laser source would also allow the range of FIR laser tuning to be extended. Based on the emission bandwidth results reported here, the generation of frequency tunable, subnanosecond FIR pulses in these laser pumped molecular lasers appears feasible.

## Figure Captions

- Fig. 1. Tunable FIR Laser System. D=Detector; W1, W2: Windows; F.M., M.: Mirrors; I1, I2: Interferometers; P1, P2: Plotters; S. A.: Signal Averaging Electronics; R: Ramp Generator; P.A.T.: Photoacoustic Transducer.
- Fig. 2. R Branch Tuning Data for CH<sub>3</sub>F
- Fig. 3. P Branch Tuning Data for CH<sub>3</sub>F
- Fig. 4. FIR Laser Emission Bandwidth

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