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## High speed frequency modulation of far infrared lasers using the Stark effect

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Electronic frequency tuning of an optically pumped far infrared waveguide laser has been achieved by using the Stark effect. Frequency modulation with a 50-kHz modulation frequency and an index greater than 1 has been observed as well as a maximum modulation frequency of 300 kHz:

We report the wide bandwidth, high index frequency modulation of an optically pumped far infrared (FIR) laser operating at the 496- $\mu\text{m}$  transition in  $\text{CH}_3\text{F}$ . The development of FIR waveguide lasers is a rapidly expanding field. This new tuning capability is expected to open up many useful applications in spectroscopy, telecommunications, and frequency standards. Of particular importance is the ability to synthesize frequencies in the far infrared. The demonstrated tuning capability should make it possible to phase-lock almost any FIR laser to a signal multiplied up from the microwave region, resulting in far infrared frequency sources with no loss of precision relative to the cesium beam frequency standard.

An initial, static frequency shift is produced by offsetting the frequency of the laser cavity from the molecular transition, introducing a certain amount of cavity pulling. The modulation is then achieved by applying an electric, or Stark, field across the laser medium. The applied field varies the effective width of the molecular transition and thereby changes the magnitude of the pulling. This effect is described later in more detail. Compared to other modulation schemes, such as electrooptic or acousto-optic modulation, this method avoids the introduction of lossy material inside of the cavity. Moreover, the Stark effect provides high speed frequency tuning, limited only by the life-time of photons in the cavity.

Figure 1 shows schematically the experimental setup. It consists of a  $\text{CO}_2$  laser system for pumping; two far infrared lasers (FIR-I and FIR-II) operating at the same transition, one of which serves as a reference, while the

other is frequency tunable; a He-Ne alignment laser; a spectrum analyzer for monitoring the  $\text{CO}_2$  lines; and a diode detector which is illuminated by the two FIR laser beams. The following details are worth noting.

The pump laser is a flowing gas commercial  $\text{CO}_2$  laser. It is tuned by a grating to the  $P(20)$  transition in the 9.6- $\mu\text{m}$  band. The output power is amplified nearly a factor of 2 by a  $\text{CO}_2$  gas discharge in an amplifying tube. The available power is 34 W at the line center of the  $\text{CO}_2$  laser. A ZnSe beam splitter divides the output into two separate beams which are directed and focused by two ZnSe lenses into the input coupling holes of the two FIR lasers. The pump powers available at FIR-I and FIR-II are 15 W and 11 W, respectively, when the length of the  $\text{CO}_2$  laser is adjusted for maximum FIR laser power.

The FIR-I laser consists of a metal tube, 3 m long with a 14-mm i.d. The input and output coupling holes are cut in flat copper mirrors and have diameters of 1 mm and 5 mm, respectively. The output coupling hole is off center with respect to the tube axis in order to give more efficient pumping and higher output power, approximately 1.5 mW for the 496- $\mu\text{m}$  transition in  $\text{CH}_3\text{F}$ . This laser is adjusted, in length, for maximum output power and serves as the reference oscillator against which the frequency of the FIR-II laser is measured.

The FIR-II laser uses a Pyrex tube, 1.05 m long with a 34-mm i.d. The input coupler is a 2-mm diam hole in a flat copper mirror; the output coupler is an 8-mm diam hole in a flat aluminum mirror. The output hole is off center from the axis of the tube and yields a maximum power, without Stark electrodes, of 2.1 mW. In the actual Stark shift measurements the internal electrodes consisted of 12.5-mm wide copper-coated adhesive tape applied over the total length of the Pyrex tube. This reduces the output power a factor of 2-4 due to the increased surface irregularities and the different boundary conditions for the various modes. The presence of the electrodes within the cavity also affected the mode structure of the laser. When the electrodes were positioned so that the static electric field was orthogonal to

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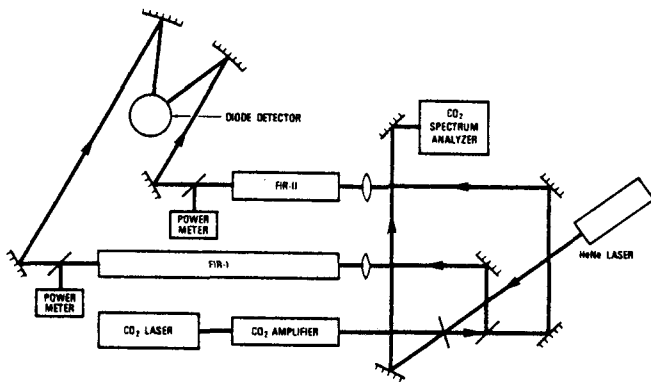


Fig. 1. Schematic diagram of system for heterodyning two far infrared (FIR) lasers. The frequency of FIR-II may be tuned by applying an electric field.

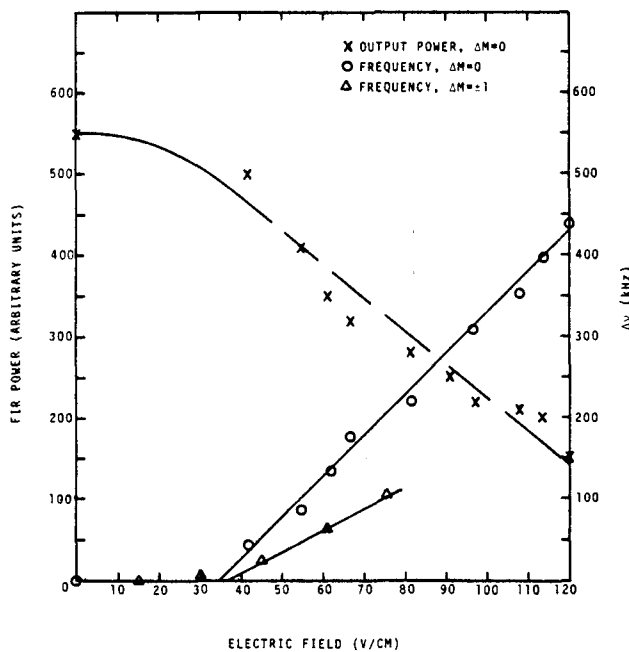


Fig. 2. Frequency shift and output power of a far infrared (FIR) laser with static electric field applied parallel ( $\Delta M = 0$ ) and perpendicular ( $\Delta M = \pm 1$ ) to the oscillating electric field.

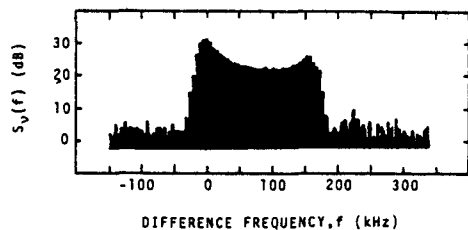


Fig. 3. Frequency modulation spectrum (spectral density of voltage) of a far infrared (FIR) laser as the result of the simultaneous application of a static electric field and a 2-kHz electric field. The vertical scale is 10 dB/division, and the horizontal scale is 50 kHz/division.

the pump polarization, different far ir cavity modes were observed polarized both parallel and perpendicular to the pump field. Stark shift measurements were performed for both polarizations.

Alignment of the CO<sub>2</sub> pump beams on the axes of the two FIR lasers is extremely important. This is true also for the angular orientation of the mirrors on these lasers. Alignment is achieved using a He-Ne laser beam, which is superimposed on the CO<sub>2</sub> beam by using a NaCl window, as indicated in Fig. 1. This also makes it possible to extract a small portion of the CO<sub>2</sub> beam to divert to a CO<sub>2</sub> spectrum analyzer.

A metal-insulator-metal diode with a tungsten whisker and a nickel post was used as a detector. Using two spherical mirrors, the two FIR beams were focused at the optimum angle on the whisker for maximum coupling to a long wire antenna.<sup>1</sup> The heterodyne signals were recorded using a power spectrum analyzer.

Static amplitude and frequency changes of the FIR-II laser were observed by first detuning the cavity of that laser from the transition line center and then applying a dc voltage to the tuning electrodes. The reference laser, FIR-I, was tuned to its transition line center. The output power of FIR-II was measured using a thin plastic beam splitter and a Golay cell, while the frequency changes were measured directly using the heterodyne signal from the two lasers. The amount of frequency tuning with applied electric field depends primarily on the degree of detuning of the laser cavity from the center of the molecular resonance. Typically the laser has been operated with an initial cavity detuning of 8 MHz, and an additional 500-kHz electronic tuning has been obtained. Over most of the range, the tuning rate is linear with the applied field. Values as high as 5 kHz/(V/cm) have been achieved. Figure 2 shows typical changes in amplitude and frequency produced by the application of the dc electric field.

Since both the amplitude and frequency of the laser change with the applied field, the application of an alternating field produces both amplitude and frequency

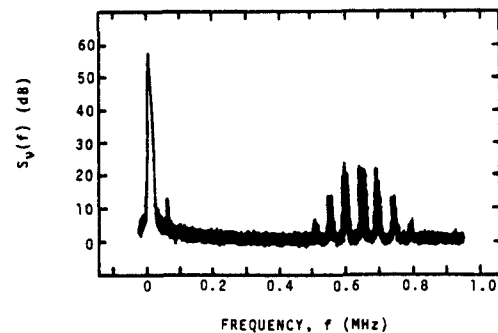


Fig. 4. Frequency modulation (spectral density of voltage) of a far ir laser modulated at 50 kHz with a modulation index greater than 1. From the left to the right are a large peak due to the local oscillator of the spectrum analyzer followed by a single amplitude modulation peak and then the carrier and six sidebands which are 90% due to frequency modulation. The vertical scale is 10 dB/division, and the horizontal scale is 100 kHz/division.

modulation sidebands. When the sidebands are small compared to the carrier, the static data of Fig. 2 may be used to estimate the expected amplitude ratio of the AM sidebands to the first FM sidebands. For the range of electric field where the frequency shift is linear, the measured sensitivities are 5 kHz/(V/cm) and 0.01/(V/cm), yielding a ratio of  $2 \times 10^{-6} f_m$ , when  $f_m$  is the modulation frequency in Hz. This implies that the AM and FM sidebands would have equal strength at  $f_m = 500$  kHz.

Figure 3 and 4 are photographs of data taken with a spectrum analyzer. They show the spectral density of the voltage fluctuations ( $V^2/\text{Hz}$ ) of the heterodyne signal. Since only the FIR-II laser is modulated, the spectra of the heterodyne signal and the FIR-II laser are proportional. The vertical scales are 10 dB/division while the horizontal scales are 50 kHz/division and 100 kHz/division, respectively. Although such a spectrum analyzer does not separate amplitude and frequency modulation, the characteristics of the spectrum can be used to determine the relative magnitudes. Figure 3 illustrates the spectrum of the laser modulated at a 2-kHz rate by the application of a 90-V/cm static electric field and a 20-V/cm alternating field. The individual sidebands are not resolved, but the spectrum has the characteristic shape of high index FM, with two well defined peaks which are separated approximately 150 kHz. The modulation index is defined as the frequency deviation (half the peak-to-peak frequency excursion) divided by the modulation frequency.<sup>2</sup> From this we conclude that the frequency deviation is 75 kHz, and the modulation index is approximately 37.5. Figure 4 shows a spectrum of the laser modulated at 50 kHz with a modulation index greater than 1. The large peak at the far left of Fig. 4 is the carrier of the spectrum analyzer which appears at a position corresponding to zero frequency difference between the two lasers. The small peak next to it is the amplitude modulation of the FIR-II laser. On the right is the spectrum of FIR-II heterodyned down to approximately 0.7 MHz by mixing with FIR-I. The carrier is in the center with three sidebands visible on either side. The first sidebands on either side of the carrier are slightly more powerful than the carrier, indicating a frequency modulation index of approximately 1.4, and are ten times more powerful than the baseband AM signal. This indicates that the heterodyne modulation spectrum is nearly pure frequency modulation with only 10% distortion due to amplitude modulation. In other experiments, a maximum modulation frequency of 300 kHz has been observed.

The  $\text{CH}_3\text{F}$  laser system was selected for study because it has a strong line at  $496 \mu\text{m}$  (600 GHz), sufficient spectroscopic data are available to characterize the energy levels, and the Stark effect is linear and well understood.  $\text{CH}_3\text{F}$  has a vibrational absorption band centered near the  $P(20)$   $\text{CO}_2$  laser line at  $9.55 \mu\text{m}$ . We observe only the stronger of the two subbranches of this absorption band,  $Q(12,2)$  ( $\nu_3 = 0 \rightarrow 1, J = 12 \rightarrow 12, K = 2 \rightarrow 2$ ), which gives rise to the  $496\text{-}\mu\text{m}$  rotational transition ( $\nu_3 = 1 \rightarrow 1, J = 12 \rightarrow 11, K = 2 \rightarrow 2$ ). The Doppler width of the rotational transition is 1.28 MHz,

and the homogeneous line width is generally accepted to be 300 kHz/Pa (40 MHz/Torr).<sup>3</sup> Our own measurements indicate that during these experiments our lasers were slightly pressure broadened.

Since  $\text{CH}_3\text{F}$  is a symmetric top molecule it has a first order Stark effect.<sup>4</sup> The rotational transitions obey the selection rules  $\Delta J = \pm 1, \Delta K = 0$ , and  $\Delta M = 0, \pm 1$ . The frequencies of the various components are

$$\nu = 2B(J+1) + \frac{2MK\mu E}{J(J+1)(J+2)h},$$

when  $\Delta M = 0$  and

$$\nu = 2B(J+1) + \frac{(2M \mp J)K\mu E}{J(J+1)(J+2)h},$$

when  $\Delta M = \pm 1$ , and  $J$  is the smaller of the two quantum numbers involved.

The relative intensities of the Stark components for different  $M$  values are  $(J+1)^2 - M^2$  when the static and rf fields are parallel ( $\Delta M = 0$ ) and  $[(J+1) \pm (M+1)](J+1 \pm M)$  when the static and microwave fields are perpendicular ( $\Delta M = \pm 1$ ).<sup>4</sup> Figure 5 shows the calculated shape of the Stark broadened transition assuming

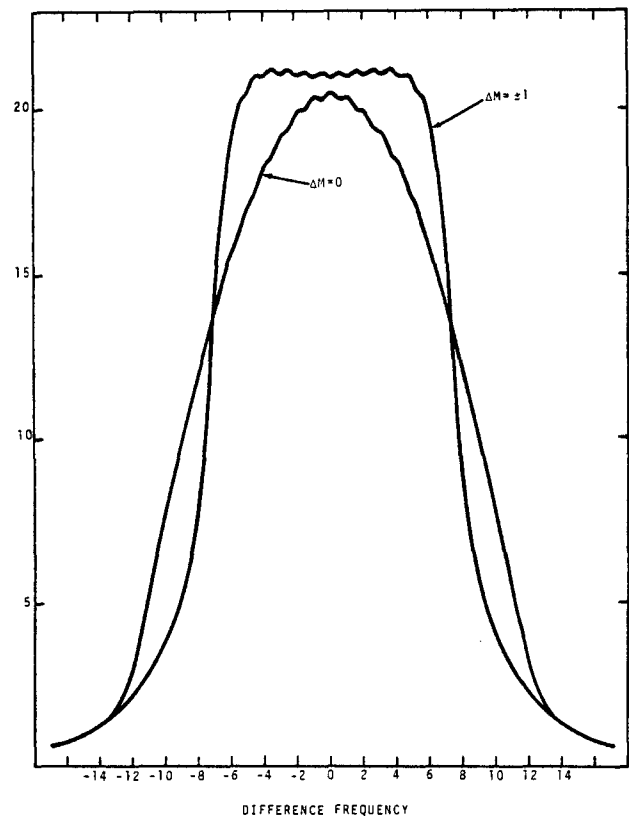


Fig. 5. The computed intensity profile for the  $496\text{-}\mu\text{m}$  transition of  $\text{CH}_3\text{F}$  for the case where the difference frequency between adjacent lines is equal to the width of a single line. The horizontal axis is in units of the homogeneous line width for zero electric field.

that the separation of neighboring components is equal to the full width of a single component and that each component has a Lorentzian shape, i.e., the line is pressure broadened.<sup>5</sup> When the appropriate constants for CH<sub>3</sub>F are used it is found that the width of the Stark broadened line is approximately 30 kHz/(V/cm) for both the  $\Delta M = 0$  and  $\Delta M = \pm 1$  transitions.

The possibility of tuning a cw CH<sub>3</sub>F laser several GHz by operating on a single Stark component was suggested a short time ago.<sup>6</sup> However, the decrease in gain which results from using fewer molecules results in a cessation of oscillation before the required electric fields are obtained.<sup>7</sup> The tuning which we have observed is, instead, the result of the Stark line broadening. Offsetting the frequency  $\nu_c$  of the laser interferometer from the center of the molecular transition  $\nu_m$  produces a static frequency shift of the laser by the effect of cavity pulling.<sup>8</sup> The magnitude of the frequency shift is determined by

$$\nu - \nu_m = [(\Delta\nu_m)/(\Delta\nu_c)] (\nu_c - \nu),$$

where  $\Delta\nu_c$  is the full width of the laser cavity and  $\Delta\nu_m$  is the full width of the transition, and the cavity is assumed to be wider than the transition. Differentiation with respect to the applied field  $E$  yields

$$\frac{d\nu}{dE} = \frac{\Delta\nu_c(\nu_c - \nu_m)}{(\Delta\nu_m + \Delta\nu_c)^2} \frac{d}{dE} (\Delta\nu_m).$$

The zero field line width of the transition  $\Delta\nu_{mo}$  has been calculated from the data of Fig. 2: The Stark frequency shift is very small until a certain critical field, approximately 40 V/cm, for which the Stark broadened line width is comparable to the transition line width. Since the Stark broadening is about 30 kHz/(V/cm) the width of the transition is approximately 1.2 MHz, which is comparable to the Doppler width. The zero field line width was fixed in these experiments since the pressure was constant throughout. The line width of the FIR-II laser cavity has been determined by measuring the frequency pulling at zero electric field which yields  $\Delta\nu_c = 17\Delta\nu_{mo}$ .

The technique of electronically tuning an FIR laser has been demonstrated to produce at least 500-kHz static tuning as well as frequency modulation with a modulation index of 1.4 and a modulation frequency of 50 kHz. Since the output line width is typically 5 kHz for an unstabilized FIR laser and better than 1 kHz for a well stabilized system, it should be straightforward to phase-lock such a laser to a reference signal. The electronic tuning which has been demonstrated for CH<sub>3</sub>F is applicable to other FIR lasers.

Depending upon the operating conditions, the Stark frequency modulation may be accompanied by significant amplitude modulation. If necessary, the magnitude of the amplitude variations may be reduced by either using an external leveler or by choosing an operating point which has a very small ratio of amplitude to frequency modulation. Tobin and Jensen<sup>7</sup> have shown that in the case of CH<sub>3</sub>F and CH<sub>3</sub>OH, at a value of electric field which should produce large frequency tuning, the slope of the amplitude vs Stark electric field curve for the  $\Delta M = \pm 1$  transitions may become zero.

Although we have not observed the same static behavior in the case of CH<sub>3</sub>F, we have been able to verify the existence of operating points where the amplitude modulation vanishes.

There is a possibility that specific molecules may be found which have very much larger Stark tunability. In particular, if a molecule which has a high rotational constant and which can be optically pumped is found, it may have a transition in the infrared with a very small  $J$  value. In this case, a FIR laser using this molecule may oscillate on a single Stark component. Since the Stark splitting is approximately proportional to  $J^{-2}$ , a factor of 100 increase in tunability could be obtained. The disadvantage of this approach is that it is likely to yield only a small number of tunable lasers in the far infrared.

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