INFRARED FREQUENCY SYNTHESIS

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The activity in infrared frequency synthesis (IFS) has been a recent development relative to the more precise work we've heard described the last few days. While it does not approach the precision and accuracy of that work in its present stage, there is reason to believe it can be improved significantly.

I'll give a brief history of some of the developments in IFS, and a detailed account of some particular measurements (those performed at NBS). Finally, I'll indicate plans and work in progress at NBS for the objective of measuring the frequency of the methane stabilized He-Ne laser, which is a candidate for the unified time and frequency standard.

A project to measure the frequency of the HCN laser at NBS was proposed by K. Evenson in late 1965. Hocker, Javan and workers at MIT apparently also started work in this area about the same time. The first slide shows the principle laser lines in the HCN laser. The 311 and 337 μm lines are vibrational-rotational transitions between these levels shown here.

Not shown here is the 373 μm line which is a pure rotational transition between $j = 9$ and $j = 10$ levels of the $04^{0}0$ vibrationals state. This is probably the longest wavelength laser line of much use.

The second slide shows a block diagram of a representative experimental arrangement for measuring the frequency of the 337 μm line of the HCN laser.

The reference point for the frequency measurement is a stable quartz oscillator whose frequency is 106.296350 MHz. An x-band klystron is phase locked to the 100th harmonic of the quartz reference (with a 30 MHz offset). An E-band klystron is in turn phase locked 30 MHz from the 7th harmonic of the x-band klystron. The output of the E-band klystron goes to a harmonic generator mixer where its 12th harmonic is compared with the 891 GHz output of the HCN laser and the approximate 30 MHz difference is measured on the spectrum analyzer. In the arrangement diagrammed here, the laser has a beam splitter output-coupling and the beam is focused into a microwave waveguide horn.

The first published value of a frequency measurements of the HCN laser came in 1967 when L. Hocker, J. Javan, D. R. Rao, L. Frenkel and T. Sullivan reported 890.7595 GHz and 964.3123 GHz for the 337 and 311 μm lines. An independent measurement at NBS reported 890.7606 GHz for the 337 line which is well within the 6 MHz gain bandwidth of this laser and represents the experimental uncertainty in determining the top of the laser line.

As one progressed to higher frequencies, it became increasingly difficult to get there with solely klystron frequency harmonics.

In general, to measure an unknown laser frequency, ν_x, one requires one or two other lasers and a klystron whose frequencies satisfy the condition:

$$\nu_x = \ell\nu_1 + m\nu_2 + n\nu_{kly} \pm \nu_{1F}.$$

ν_1 and ν_2 are basis laser frequencies where ℓ, m, and n are harmonic numbers with m and n allowed both positive and negative values. Wavelength measurements are sufficiently good to determine the harmonic numbers before hand. The uncertainty in wavelength is compensated for by having a tunable klystron and performing some frequency searching initially.

The third slide summarizes some of the measurements made to date. This list makes no pretense at being exhaustive. Instead, it is a compromise between reasonable basis frequencies and including references to different workers in the area. The initial measurement on the HCN laser I've already mentioned. The first reference on the slide shows the frequency of three HCN lines as reported by Hocker and Javan in 1967. (They actually reported more than these, but these are used in subsequent measurements.)

The three here are 373, 337 and 311 μm. The 337 μm line at 0.8907607 THz is obtained from the 12th harmonic of a 74.2 GHz klystron and the frequencies of the 373 and 311 μm lines are obtained by mixing the third harmonic of 28.7 and 26.5 GHz respectively. These were all made using a tungsten catwhisker on a silicon chip for a diode. A sufficiently strong signal comes from the diode that this measurement is very routine now and the signal has been used to servo the HCN laser and make it oscillate at a constant frequency.

Progressing to the next block, we see the 190 μm line of DCN. This line as well as the 195 and 204 μm lines were measured by Hocker and Javan in 1968. Frenkel, Sullivan, Pollack and Bridges had measured the frequency of the 118 μm line in the water vapor laser in 1967. In 1969 Hocker, Sokoloff and Javan measured the 84 μm line of the D_2O laser. All of these measurements were made with a tungsten on silicon diode.

These lines in this block are included mainly for historical reasons. In general, to synthesize a frequency, one would like to keep the number of basis lasers small for practical reasons. If one has an HCN laser, it is much easier to synthesize the 190, 118 or 84 μm lines than it is to build a new laser. As a practical matter, only klystron harmonics up to the 6th (assuming available klystrons up to 75 GHz) would be required, since one could always increase the harmonic number of the HCN laser by one and subtract klystrons harmonics.

The next important step in IFS came in 1969 when Daneau, Sokoloff, Sanchez and Javan reported harmonic generation and mixing in a tungsten on nickel diode.

The MOM diode, along with several other developments to be mentioned later, led to the next measurement on the slide by Evenson, Wells, Matarrese and Elwell at NBS. This is the 78 μm line of the water vapor laser whose frequency is 3.821775 THz. This was obtained from the 6th harmonic of 0.891 THz minus the 2nd harmonic of the 0.804 THz line plus 0.029 THz from a klystron.

This line has a desirable intermediate frequency to be used as a basic laser, however it is somewhat inconvenient to work with in that it is absorbed 15-30 db/meter in air. I should admit that we did not set out to measure this frequency, but rather discovered it inadvertently while searching for the 28 μm water vapor line. This accident was due to the fact that the klystron frequencies are nearly the same for both the 78 and 28 μm lines. The 28 μm line, by comparison with the 12th harmonic of .891 THz plus 0.029 THz from a klystron, was determined to oscillate at 10.718073 THz.

Both the 28 and 78 μm lines were used as basis frequencies to measure P-18 and P-20 lines in the CO_2 laser a few months later. The harmonic number, 3 for 10.718 THz and -1 for 3.8 THz gave a much lower order mixing number than the 12 and 13 for the previous water vapor frequency measurements. Much closer to 3 times the 10.7 THz line are the R-30, 32 lines in the 9.3 μm band of the CO_2 laser, as determined by Daneau, Sokoloff, Sullivan and Javan in a frequency measurement utilizing a pulsed CO_2 laser. The difference frequencies around .022 THz were again from a klystron.

Based upon values from these four measurements and his own work on difference frequencies, Chang published values for frequencies for all the CO_2 laser lines in the 10.6 and 9.2 bands.

One of these (R-30) was used as the basis frequency in measuring the 5.2 μm CO laser line at 58.0234341 THz. To my knowledge, this represents the highest frequency yet synthesized.

We can now go back and look at some second generation experiments. I'm referring mainly to McDonald and colleagues (Risley, Cupp and Evenson) exciting work using the Josephson junction as a mixer. A published report early this year described beating the 84th harmonic of an x-band klystron with the 0.891 THz line and getting excellent signal to noise. References 10 and 11 here have yet to be published. A 3.8 THz signal has been synthsized using the 4th harmonic of the .964 THz m HCN line plus the third harmonic of an x-band klystron. Less than 3 weeks ago, they observed a beat between the 3.8 THz line and the 401st harmonic of an x-band klystron.

I'd like to go back now and describe some of the details involved in the measurements at NBS. The first departure from the experimental setup shown earlier is that an open structure diode as shown in the next slide is used instead of a diode at the end of a horn. This permits one to focus several laser beams onto the diode simultaneously. The post here is silicon or nickel and the catwhisker is initially 25 μm diameter tungsten and, after etching, may be 15-20 μm m in diameter. The kink is a spring to provide contact pressure against the silicon base. For calculations, the length L is from the point to the start of kink.

It was suggested by Elwell that antenna theory might be applied in order to obtain the best coupling to the whisker which acts as an antenna for the laser radiation. The fifth slide shows the radiation pattern from an antenna. The expression for the angle between the first maxima and the antenna depends on the radiation wavelength and length of the antenna.

$$\Theta = \cos^{-1}\left(1 - \frac{0.371\,\lambda}{L}\right)$$

Matarrese and Evenson investigated coupling as a function of angle and found good agreement with theory as shown on slide six where the length is 7 λ with $\lambda = 337$ μm. Shown here in the solid line is the rectified diode voltage due to the laser radiation as a function angle between antenna and incident radiation direction. The dashed line is the theoretical prediction and reasonable agreement is evident.

Contact between the antenna and the diode was made by running the base into the antenna. Efforts were made to keep the diameter of the antenna small compared to a wavelength and if the base was run in too hard, the antenna turned into a fish hook. Even with a differential screw arrangement, it was difficult to make a light contact. The diode is mounted on a 3-dimensional manipulator which is used to position the diode with respect to the focal point of the output laser lens. A lever arm is attached to the micrometer on the differential run-in screw to facilitate lighter contact of the diode.

The improved antenna coupling alone was not sufficient, as our early attempts to measure the frequency of the water vapor laser were unsuccessful.

Based on the assumption that power into the harmonic went as $\frac{1}{n^2}$ and the fact that we were looking for the 12th harmonic, it was decided to lengthen the lasers from 4 m to 8 m to get as much power as practically possible. This dictated a move to a larger lab where the experiments were eventually successful. The next slide (seven) shows an 8 m HCN laser and two 8 m H_2O vapor lasers. The device on the left is Don McDonald's dewar containing the Josephson junction.

Concurrent with lengthening the lasers was another development for getting more power from them. This improvement was a Michelson interferometer output coupling scheme. The new laser design is shown in the following slide.

Invar spacers minimize the drift between the end mirrors. Let's neglect the side mirror initially. A 16 m radius curved mirror is at one end and a flat mirror at the other end complete a folded-confocal laser arrangement. A dielectric beam splitter and polyethylene output lens are shown. Part of the radiation propagating from mirror C is deflected toward mirror A. (It's field and power we'll call E_o and P_o). The rest passes through the beam splitter, is reflected from mirror B and part of this is subsequently deflected from the beam splitter where it passes thru the output lens and is focused down to a point about 20 cm from the laser. Mirror A is then aligned so that the radiation reflected from it is focused at the same point. It is then possible by translating mirror A to vary the relative phase of the radiation reflected from the two mirrors. The resulting field can be made to vary from 0 to 2 E_o. The resulting power is then 4 P_o.

In order to reflect the beam from the laser, a thin polyethlyene film is glued to the beam splitter frame. The thickness is chosen such that one has constructive interference between the reflected beams from the front and back surfaces of the film. The thickness is an odd multiple of t where

$$t = \frac{\lambda}{4}\left(\frac{1}{n^2 - 1/2}\right)^{-1/2} \text{for} \quad \Theta_i = 45^\circ$$

(θ_i is the angle of incidence and n is the index of refraction)
Slide nine shows the power output from an HCN laser as a function of Michelson side mirror position. The 180° points correspond to destructive interference. Halfway between at the constructive interference point, the coupling is a maximum, however so much power is being coupled out that the field in the laser has decreased and the net coupling is decreased. The parameter is discharge current in the laser.

This Michelson coupling arrangement has several advantages over hole coupling schemes. Those relating to frequency measurements are:

1. The beam is not badly diffracted leaving the laser. (The output lens can be replaced by a window to allow use of the laser beam some distance away.
2. Four times as much power can be extracted as from a simple beam splitter.
3. The output coupling is easily varied.
4. The output beam is linearly polarized. This permits good coupling to the diode antenna.

These three factors, 1) the increased laser power due to longer laser, 2) the improved output coupling scheme, and 3) the application of long antenna theory were combined with the use of the tungsten on nickel diode and used in the H_2O vapor and CO_2 laser frequency measurements.

The next slide (ten) shows a block diagram for the H_2O vapor laser frequency measurement. The E-band phase locked chain which was shown in the first slide was used to make a simultaneous measurement of the HCN laser frequency, by splitting off a portion of the HCN beam. The HCN main beam goes to a diode along with the water vapor frequency at 29 GHz. The difference frequency is measured on a spectrum analyzer. Not shown here are adjustable mirror arrangements which were used to bring the radiation into the antenna at the appropriate angles. By slightly adjusting these mirrors, while monitoring the rectified voltage output, one could optimize the coupling of radiation from each individual source. After this optimization of coupling, there was still an uncertain factor in the experiment, namely was the diode contacted suitably? Only about 1 in 10 or 20 were. We finally arrived at a criterion to determine whether to proceed with the experiment or to contact a new diode.

In addition to good rectification, all diodes which were used successfully in frequency measurements exhibited a huge 18 MHz intermode beat note from the water vapor laser as shown on the slide 11a. The dispersion of the spectrum analyzer is 200 KHz/cm and an attenuation of 60 db was required to bring the top of the signal on scale. Mixing experiments were continued only after this criterion was met; i.e., a beat note that resembled this.

For comparison, note the beat note between the 28 μm water vapor line and the 12th harmonic of 0.891 THz as shown on the slide 11 b. (Vertical sensitivity is the same for the 2 slides.) One may determine that the beat note is the one desired by independently tuning the frequencies of both lasers and the klystron by known amounts and ascertaining that the beat note shifts a correspondingly correct frequency for each test.

In the experimental arrangement in the lab for measuring the CO_2 laser frequencies, the diode antenna is turned the proper direction from the H_2O output beam for optimum coupling and a mirror reflects the CO_2 laser beam in at the proper angle. If all other factors were equal, one would expect a considerably larger beat note here than for the H_2O measurements since the mixing order is 5 instead of 13. The beat note is larger but not as large as one would expect, as shown on slide 11c 50 KHz/cm dispersion.

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This indicates some of the factors involved in making a measurement of an unknown laser frequency. An unmeasured frequency of great interest is that of the methane stabilized HeNe laser at 88 THz. Slide 12 summarizes some possible schemes for measuring the 88 THz line. These are listed in order of decreasing complexity and probably increasing difficulty.

Experiment A is a possible scheme using a bulk material for the harmonic generation. The top line of this experiment is presently being investigated by G. Day of NBS. The output from a pulsed CO_2 laser is focused into two seleium cyrstals which my be oriented independently with respect to the propagation axis. The frequency is doubled in the first crystal and 2ω signal combined with the fundamental in the 2nd crystal to give an output at 3ω. To date he has detected 3ω signals at about 150 μwatts. The 3ω signal plus 88 THz and a 48 GHz signals are focused on an indium arsenide photo diode and he is currently searching for the beat note.

Experiment B would be similar with the harmonic generation and mixing both occurring in a MOM diode. The basis laser frequencies are the same for both schemes.

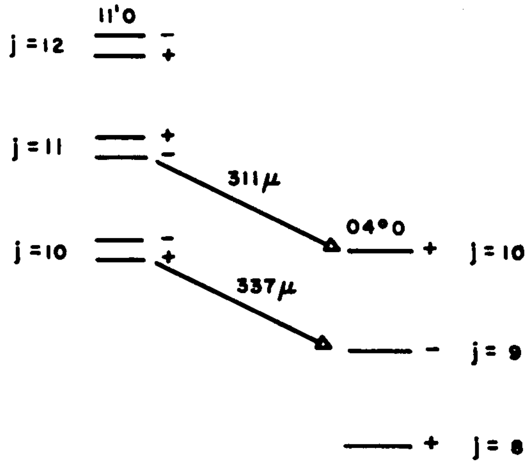
Experiment C is currently being investigated by K. Evenson. He is using a MOM diode with a 2.5 μm tungsten whisker for the antenna and hoping to mix the 8th harmonic of 10.7 plus the 3td harmonic of 0.891 minus 40 GHz from a klystron for a near coincidence. Again, due to the high order mixing, this may be a difficult experiment.

The high order mixing is considerably less of a problem with the Josephson junction. Experiment D indicates one possible scheme. The 8th harmonic of 10.7 could be combined with the 100th of a 26.3 GHz klystron for a coincidence. The 10.7 could possibly be obtained from the 1000th harmonic of x-band. This latter experiment is currently being attempted by D. McDonald and colleagues.

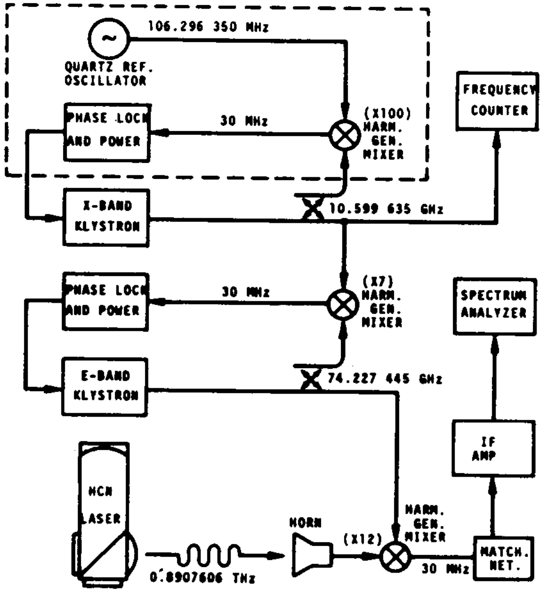
Experiment E would also be a possibility with about 100 harmonics each for x-band to .891 and 0.891 to 88 THz.

Experiment F is included for those who think big. I believe Don Halford first suggested this step.

Back to the realm of higher probability. An accurate measurement would require simultaneous measurements of the basis lasers or stabilized lasers which have been carefully characterized. Work is in progress at NBS to stabilize those lasers which are expected to be used in the 88 THz measurement. R. Petersen and B. Danielson are working a saturation absorption scheme for stabilizing the CO_2 laser. My efforts recently have been on phase-locking an HCN laser. We hope to start a water vapor laser stabilization effort soon. At present, one needs a huge improvement in the S/N of the (12 x 0.891) - 10.7 beat note or an as yet undiscovered absorption with which to stabilize the water vapor laser.



Slide 1



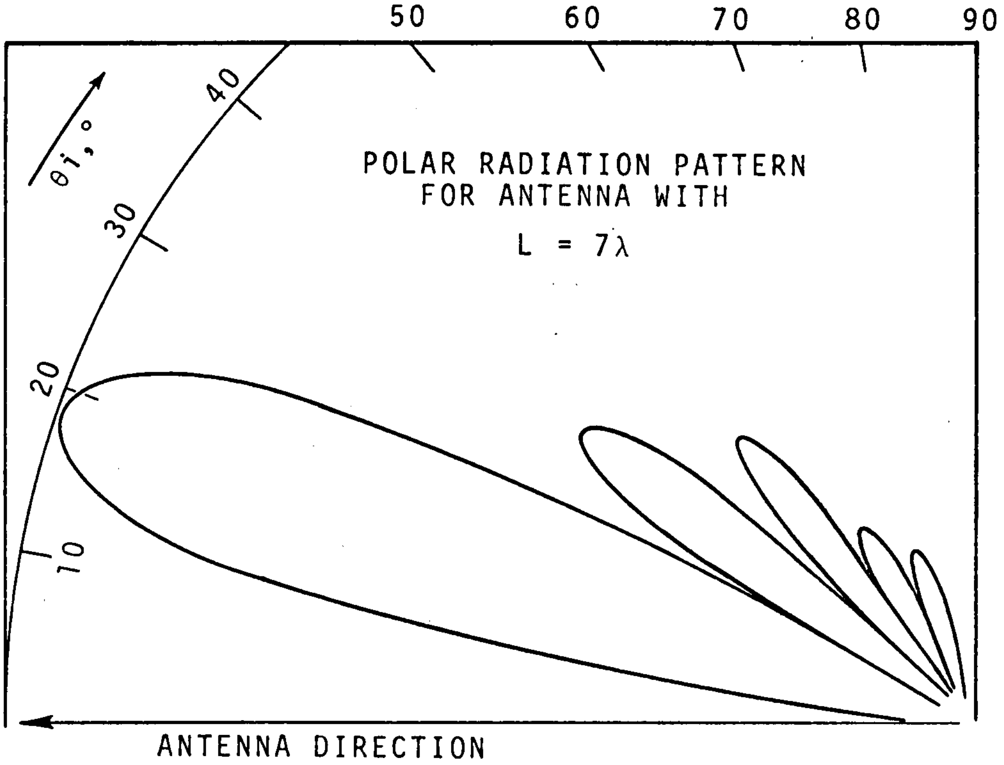
Slide 2

Results of Some Completed Synthesis Experiments

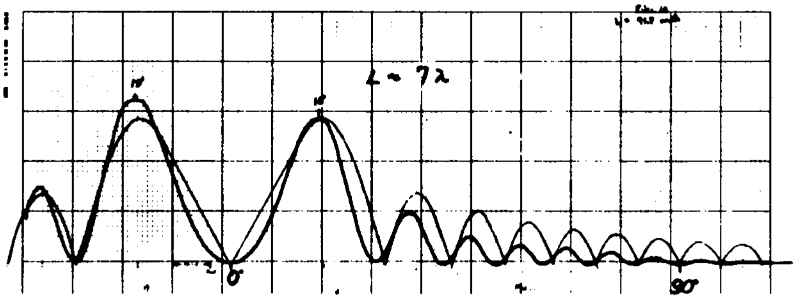
$$\nu_x = \ell\nu_1 + m\nu_2 + n\nu_3 \pm \nu_{1F}$$

Ref	ν_x in THz	Laser	λ in μm	basis laser 1		basis laser 2		basis klystron		type of mixer
				ℓ	ν_1	m	ν_2	n	ν_3	
1) HJ 1967	0.8047509	HCN	373	1	0.891	0		-3	0.0287	MOS
1)	0.8907607		337	0		0		12	0.0742	MOS
9) MRCE 1971								84	0.0106	JJ
1)	0.9643134		311	1	0.891	0		+3	0.0265	MOS
3) HJ 1968	1.578279	DCN	190	0		0		22	0.0708	MOS
2) FSPB 1967	2.527953	H_2O	118	0		0		17	0.148	MOS
4) HSJ 1969	3.557143	D_2O	84	0		4	0.891	1	0.0059	MOS
5) EWME 1970	3.821775	H_2O	78	6	0.891	-2	0.805	3	0.029	MOM
10) MRCE 1971				0		4	0.964	3	0.0089	JJ
11) MRCEA 1971				0		0		401	0.0095	JJ
5)	10.718073		28	0		12	0.891	1	0.029	MOM
6) EWM 1970	28.306251	CO_2	10.6	3	10.718	-1	3.821	-1	0.027	MOM
6)	28.359800	CO_2	10.6	3	10.718	-1	3.821	1	0.026	MOM
7) DSSJ 1969	32.134269	CO_2	9.3	3	10.718	-1		-1	0.020	MOM
7)	32.176084	CO_2	9.3	3	10.718	0		1	0.022	MOM
8) SSOJ 1970	58.024341	CO	5.2	2	29.011	0		1	0.002	MOM

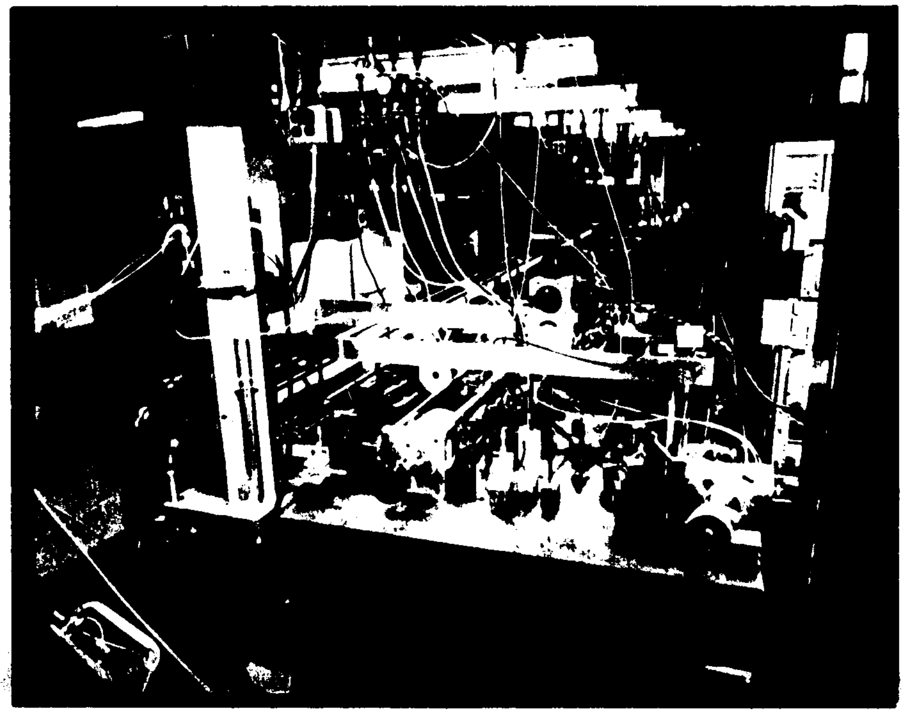
Slide 4



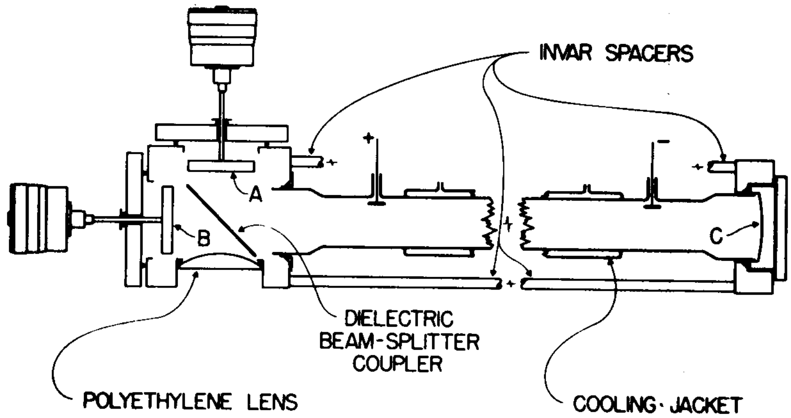
Slide 5



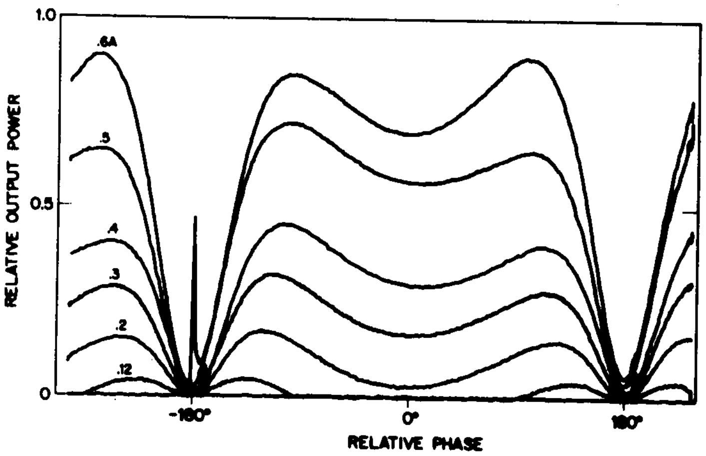
Slide 6



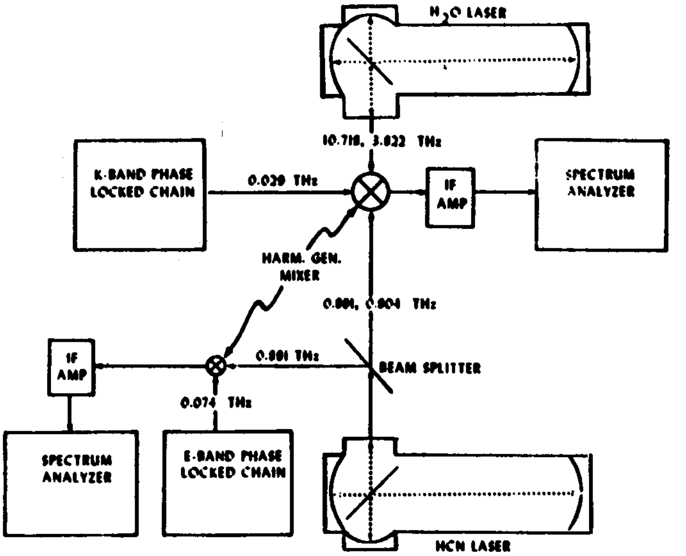
Slide 7



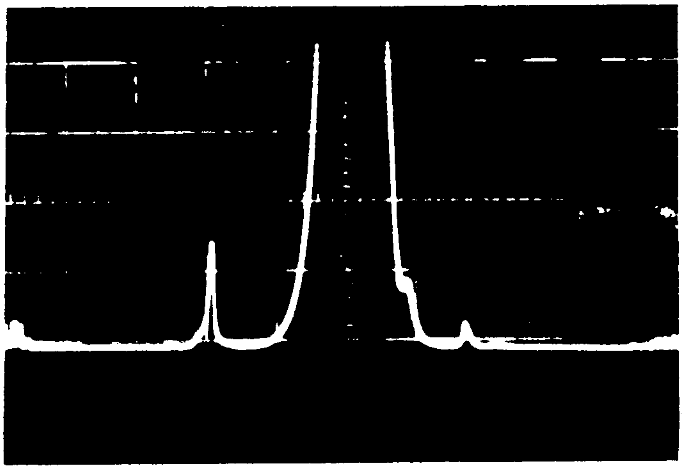
Slide 8



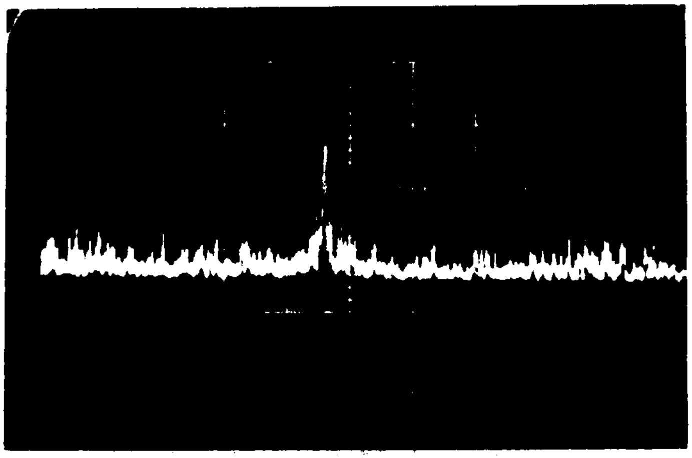
Slide 9


H$_2$O LASER
10.718, 3.822 THz
K-BAND PHASE LOCKED CHAIN
0.029 THz
IF AMP
SPECTRUM ANALYZER
HARM. GEN. MIXER
0.891, 0.964 THz
IF AMP
0.891 THz
BEAM SPLITTER
0.074 THz
SPECTRUM ANALYZER
E-BAND PHASE LOCKED CHAIN
HCN LASER

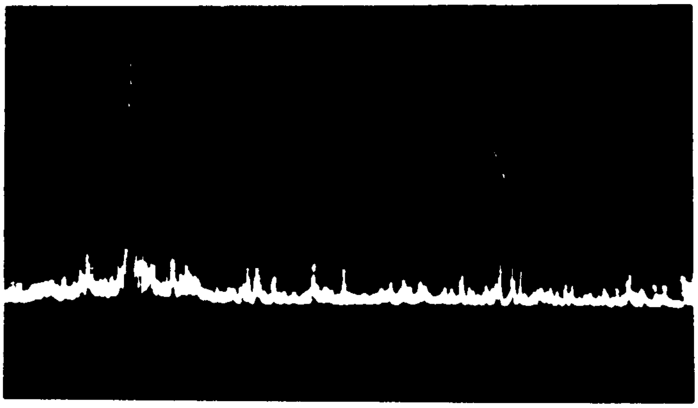
Slide 10



Slide 11a



11b



11c

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Some Possible Schemes for Synthesizing 88 THz

$$\nu_x = \ell \nu_1 + m\nu_2 + n\nu_3 \pm \nu_{1F}$$

Expt	ν_x Freq. in THz	Type Laser	Wavelength in micrometers	Laser 1 ℓ	Laser 1 ν_1	Laser 2 m	Laser 2 ν_2	Klystron n	Klystron ν_3	Mixer
A, B	88.376202	HeNe*	3.39	3	29.443	0		1	0.0487	Bulk, MOM
	29.442509	CO_2	10.2	3	10.718	-3	0.891	-3	0.0394	MOM
	10.718073	H_2O	28	0		12	0.891	1	0.0290	MOM
	0.8907606	HCN	337	0		0		12	0.0742	MOS
C	88.376202	HeNe*	3.39	8	10.718	3	0.891	-1	0.040	MOM
	10.718073	H_2O	28	0		12	0.891	1	0.029	MOM
	0.8907606	HCN	337	0		0		12	0.0742	MOS
D	88.376202	HeNe*	3.39	8	10.718	0		100	0.0263	JJ
	10.718073	H_2O	28	0		0		1000	0.0107	JJ
E	88.376202	HeNe*	3.39	0		99	0.891	4	0.0469	JJ
	0.8917606	HCN	337	0		0		100	0.0089	JJ
F	88.376202	HeNe*	3.39	0		0		10000	0.0088	JJ

*methane stabilized