Closing the Far-Infrared Gap

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The coherent spectrum has been completed from visible to submillimeter regions, thanks to a molecular system that causes unlikely substances to lase. For instance, would you believe 12 c-w lines from water?

THE GENERATION of coherent radiation in the far infrared has closed the final gap in the spectrum between the visible and the submillimeter regions.

Until recently, engineers approached this region from the microwave side with only limited success, using free-electron devices and frequency-multiplication schemes. But submillimeter-wave tubes were too expensive, incoherent sources were too weak, and bolometer detectors were too slow.

In the past five years, however, powerful new tools have become available. With these, an experimental lasing system has been built that can be used throughout the entire gap region, providing frequency selectivity and mode control.

The principal new tool was the molecular gas laser; its advent in 1964 provided an inexpensive, intense coherent source covering the difficult range between 0.3 and 774 microns with several hundred lasing lines, more or less uniformly spaced. Peak powers ranged from milliwatts to kilowatts for the various lasing lines.

A year later, the French firm CSF built its carcinotron tubes, which operate at as few as 300 microns. Hence the gap was overlapped by quantum and classical electronic sources by some 474 microns.

Semiconductor sources are now approaching one-millimeter wavelength with fundamental frequencies, and submillimeter wavelengths with harmonic frequencies. These devices include Gunn, LSA and tunnel diodes.

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Fast, semiconductor photoconductivity detectors have been developed, which operate from the visible out to eight millimeters. Microwave semiconductor detectors have been pushed down to the order of 100 microns with reduced sensitivity, and very fast, room-temperature electric tunneling detectors can now cover the range from direct current to the order of 10 microns.

Furthermore, sufficient power now exists to study the electro-optic effect out to the order of 120 microns.

Figure 1 shows several molecular lasers and the spectral ranges in which they yield lasing lines. The two-atom

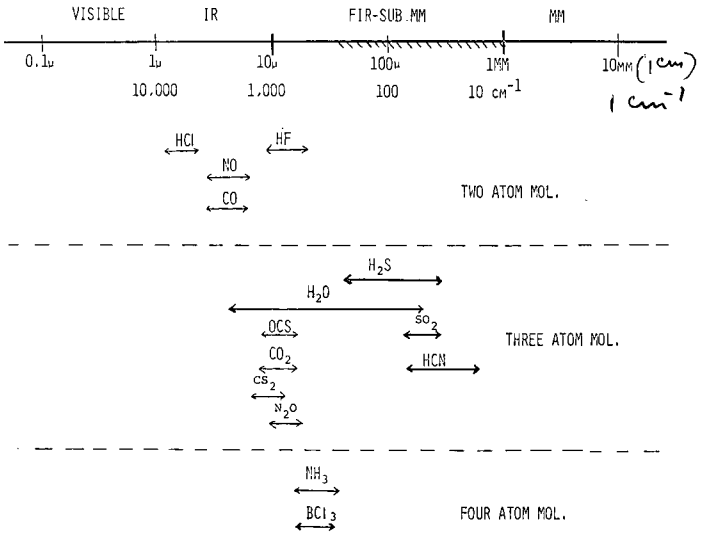

Fig 1 *Several molecular lasers and the spectral ranges in which they yield lasing lines*

molecules HCl, HF, NO, and CO are seen to yield lines in the range 1 to 23 microns. The three-atom molecules CS_2, CO_2, OCS, H_2O, H_2S, SO_2, HCN, and N_2O yield lasing lines from 7 to 774 microns for the broadest coverage of any molecular system. Finally, only two four-atom molecules, NH_3 and BCl_3, have been found to lase at this time, their lines falling in the range 16 to 33 microns.

If one were to include isotopes the list would be longer, but the spectral range of the lasing lines would be about the

same. It may be of importance that no five-atom molecule has been found to lase, probably because a sufficiently selective pumping system has not been found to overcome increased relaxation processes that exist in these molecules.

Only the two original molecules, H_2O and HCN, plus the two new molecules, H_2S and SO_2, which have been found to lase within the last seven or eight months, are here considered since they are the only ones which yield lines between 100 and 1,000 microns.

Water Yields Lasing Lines

The number of lasing lines seen thus far in these molecules is very numerous. Water vapor, for example, oscillates on more than 100 pulsed lines and over a dozen

H_2S				H_2O			
λ (Air)	τ Delay	τ Duration	Rel. Int.	λ (Air)	τ Delay	τ Duration	Rel. Int.
96.38	25 μsec	40 μsec	3	55.07	2.8 μsec	2.4 μsec	30
103.3	30	50	125	57.66	3.4	3.0	22
108.8	30	100	7	67.13	3.6	2.8	32
116.8	20	60	4	73.34	5.2	2.8	1
126.2	30	20	2	78.06	3.0	5.0	0.1
129.1	20	40	4	78.27	2.2	5.8	0.6
130.8	20	20	1	78.41	2.2	6.8	20
135.5	20	60	2	78.57	2.4	7.6	1.6
140.6	25	30	10	78.67	2.5	6.5	0.2
162.4	30	30	560	78.93	2.3	5.0	0.4
192.0	20	60	20	79.07	2.2	5.2	6.2
225.3	25	60	1000	85.50	5.8	6.2	6.0

H_2S				H_2O			
λ (Air)	τ Delay	τ Duration	Rel. Int.	λ (Air)	τ Delay	τ Duration	Rel. Int.
33.47	30 μsec	30 μsec	0.6	33.02	3.3 μsec	5.7 μsec	7500
33.64	10	40	480	35.81	4.3	4.7	900
49.62	20	40	1	36.58	5.0	6.0	1200
52.40	30	40	60	37.82	4.0	1.0	-
56.84	25	10	60	38.06	4.8	1.7	-
60.29	25	40	200	39.66	3.0	4.9	5.4
61.53	25	100	1000	40.60	3.6	0.4	74
73.52	10	40	0.2	45.50	5.6	3.4	72
80.50	25	40	220	47.22	2.6	5.4	150
83.43	30	30	0.1	47.44	5.0	8.0	960
87.47	25	100	1300	47.65	2.2	1.6	132
92.00	20	40	0.1	48.68	4.6	4.4	94

Fig 2 *Some lasing lines for selected substances*

continuous-wave lines from 7 to 220 microns. More than 24 lines have been obtained from H_2S in the range 33 to 225 microns, and I am sure more will be found. There are over 30 lines available from HCN. However, the SO_2 molecule has yielded only five lines thus far.

A partial listing of lasing lines is provided in Figure 2.

The time behavior of H_2O and H_2S, even though the two molecules have similar molecular structure, is seen to be quite different. The lasing signal from H_2O essentially occurs during the five-microsecond current pulse driving the discharge, while for H_2S there is a delay in the order of 30 microseconds before lasing occurs, and the lasing pulse lasts up to 100 μsec on several lines.

This general time of the lasing signals from the four molecules H_2O, H_2S, HCN, and SO_2, is shown if Figure 3. If one were to use these molecules in some sort of pulsed communications system or radar, there would be a problem to activate the laser pulse precisely and to achieve control-

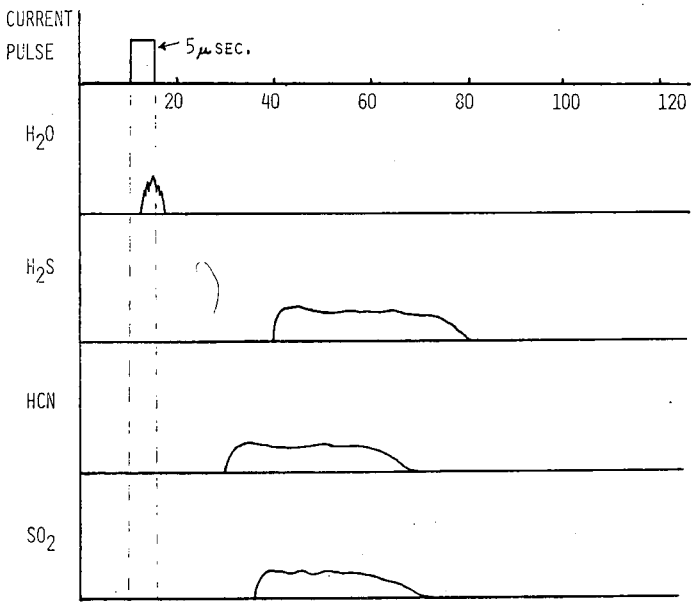

Fig 3 *Time behavior of laser signals*

lable short pulses to take advantage of the high frequency.

The detailed time behavior of two lines, 78.41 and 118.65 microns in H_2O, is shown in Figure 4. There is fairly rapid spiking taking place on a somewhat regular basis. It could be the beat note between modes, or it could

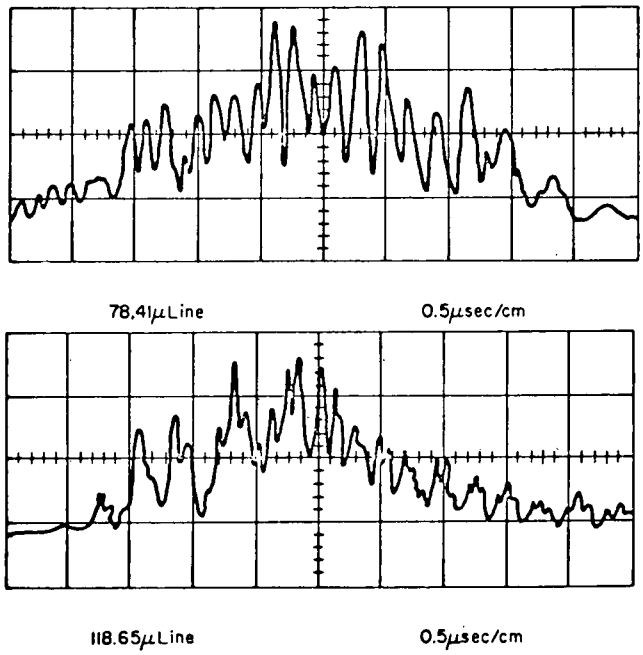

Fig 4 *Emission lines for water vapor at 118.65 and 78.41 microns*

be caused by competition between lasing lines. The 78.455 line is known to compete with the 79.106 and 115.42 lines while the 118.65 line competes with the 47.251 and cascades with the 47.693 micron line.

Excitation Techniques

The time-delay behavior of the laser signal suggests that different excitation and/or relaxation processes and times are at play in the four molecules. Excitation of the lasing specie could be achieved by electron impact, dissociation, or recombination of chemical bonds, vibrational transfer, etc.

To illustrate the time behavior of a molecular laser where the excitation mechanism is unknown, consider the molecules OCS, CO, and HF. The OCS molecule[1] is interesting since, in a gas discharge, it can be dissociated into excited CO*, which lases in the 5- to 6-micron range, while the excited CO* can pump the OCS through vibrational transfer to yield OCS* excited, which lases around 8.4 microns. The CO lasing signal begins some 20 to 60 μsec after the 1-μsec current pulse driving the discharge and lasts on the order of 30 μsec. The OSC lasing signal begins earlier at 11 μsec after the current pulse and lasts for only 4 μsec.

In the case of chemical excitation[2], as for example H_2 plus the freon CF_4, lasing starts 6 μsec after the current pulse and lasts for 5 μsec. Here the lasing lines occur in the 10- to 20-micron range.

The longest delay that we have seen in a molecular laser is in an N_2O discharge, where the lasing signal waits for a millisecond and then lases for several milliseconds. The gain and saturation parameter of the H_2O, H_2S, SO_2, and HCN lasers have been studied very little. Figure 5 gives some data on several H_2O lines that have been measured in our laboratory along with some data on helium-neon and CO_2 for comparison. It is seen that most of the H_2O lines have a

H_2O		He-Ne			CO_2		
λ	α_0	λ	α_0	S_0	λ	α_0	S_0
23.36	3.1 db/m	0.6328	0.1 db/m	-	10.6	.3 db/m	22 W/cm^2
26.66	0.57	1.1523	0.5	6 mW/cm^2			
27.97*	2.7	2.0261	4.0	-			
33.03	1.0	3.3913	26.0	-			
35.84	0.78	3.507	50.0	-			
39.70	0.57						
47.25	4.0						
47.47	1.3						

* $S_0 \sim 100$ W/cm^2

Fig 5 *Laser-gain comparisons*

pulsed gain similar to the weaker lines in He-Ne and the CO_2 lines. The value $S_0 \sim 100$ watts per square centimeter for the 27.97 H_2O line has been estimated. We have measured pulsed powers on this line in the 2- to 4-kilowatt[4] range, which indicates a large saturation parameter.

Laser System and Techniques

A schematic of a typical "Illini" molecular laser system is shown in Figure 6. For the laser tube we can use 10.2-cm ID Kimax brand glass pipe which is flanged together with "O" rings.

A far-infrared molecular laser is generally a long device, since the gain per unit length of the lines is low. As the laser gets longer, the beam spot size increases requiring larger and larger mirrors to keep the diffraction loss low. Representative values of the spot radius r for the wavelength limits 100 and 1,000 microns are given. It is seen that this system will not quite reach 1,000 microns, since at this point the beam radius, 5.74 cm, exceeds the laser tube radius, 5.10 cm.

We use a dual 5C22 line-type modulator with a 12.5-ohm impedance. This allows a two-meter section of the laser to be driven with pulse currents as large as 800 amperes. Typical pulse lengths used are 3 to 15 μsec.

The optical resonator indicated uses a spherical mirror of radius 10 to 100 meters and a flat mirror containing a coupling hole. This resonator does not contain any particu-

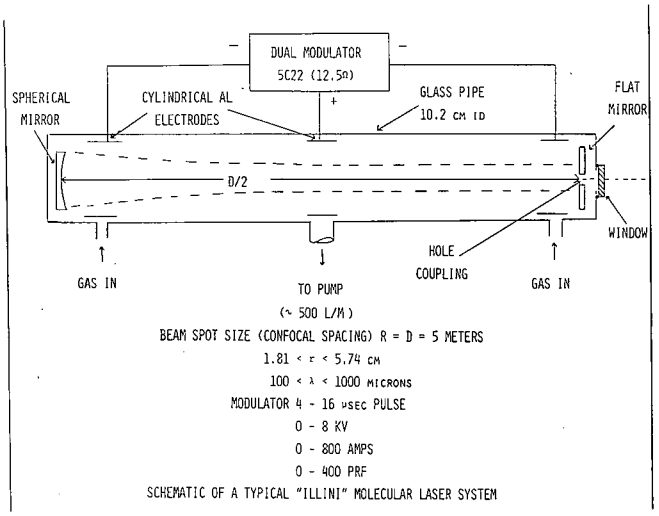

Fig 6 *Typical 'Illini' molecular laser system*

lar frequency selective characteristics other than its own resonances so that many molecular lasing lines can oscillate simultaneously.

Simple Gratings for Single Line Studies

A simple scheme to select one lasing line at a time is the grating-resonator[5] shown in Figure 7. Here the flat mirror has been replaced by a metal reflection grating blazed to the appropriate wavelengths of interest.

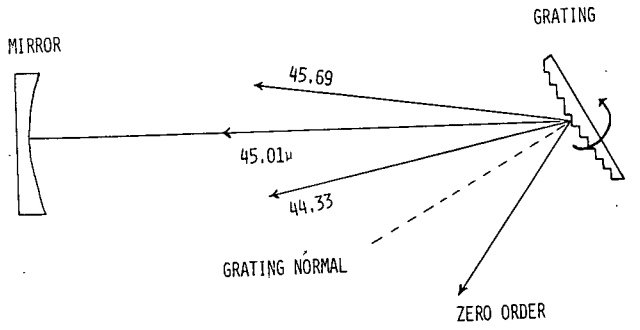

Fig 7 *Frequency-selective resonator with single grating*

The selectivity of the grating can be estimated as follows. Assume the Q of the resonator is reduced appreciably by moving the beam spot a distance S from the mirror center. Then the corresponding angular displacement $\Delta\theta$ is given by:

$$\frac{D}{2} \Delta\theta = S$$

where D/2 is the mirror-grating separation.

Using the grating equation, one can obtain

$$\Delta\lambda = 2 d \cos\theta \Delta\theta$$

Hence, for angles near the blaze

$$\frac{\Delta\lambda}{\lambda B} = \frac{2S}{D} \cot\theta_B$$

Coupling out of this resonator can be through a hole in the spherical mirror or using the zero order (Snell's-law angle) of the grating.

To study cascade-competition effects in molecular lasers, a dual grating-resonator,[6] shown in Figure 8 can be used. Consider the simple cascade situation drawn in the figure. If laser transition 1 is not allowed to oscillate, transition 2 will not oscillate, or if resonator 1 is tuned to transition 1 and resonator 2 to transition 2, then transition 2 will get stronger. The opposite situation will hold for the competition case drawn in the figure.

For measuring pulsed power, we use a fast detector to

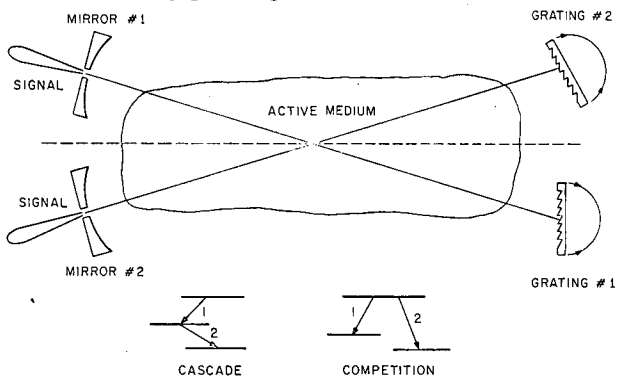

Fig 8 *Resonator with double grating*

determine the signal wave shape, from which the duty cycle can be calculated, and a simple liquid calorimeter.[7]

Using alcohol for the liquid, this calorimeter had a sensitivity of 0.001 volt per watt. The laser power is determined from the DC power and polyethylene window transmission at the wavelength of interest.

On some measurements made on the 28 micron line of H_2O in the summer of 1967, we obtained 2 kilowatts of pulsed power measured 4 inches from the output laser mirror. This distance number is important since the 28-micron line is strongly attenuated in humid aid.

Molecular Lasers in the Sky

It has been known for many years that various molecules (CN, CH, C_2, NH, OH, ScH, MgH) exist on the disk of the sun. Also molecules (CN, CH, T, O, MgH, S, H, AlH, ZrO, ScO, CrO, etc.) have been identified in stars. The molecules CH, CH^+, and CN have been observed in interstellar space.

In 1963, H. H. Barrett[8] and his colleagues at MIT detected absorption in OH radicals in interstellar space, and in 1965 a group at Berkeley and one at Harvard simultaneously detected emission from OH radicals at 1.665 and 1.667 GHz from the radio source W49.

An energy-level diagram of the OH radical, showing the hyperfine splitting of the ground state, is given in Figure 9(A). The energy of the levels is labeled in GHz instead of wave numbers for convenience. Normal spontaneous emission intensities of the four transitions are in the ratios of 1:5:9:1 as indicated.

However, when the emission from W49 was measured, the ratio of intensities of the 1.665 to 1.667 line was 3 to 1 instead of 5 to 9. Also, the effective radiating temperature was estimated to be in excess of 10^{13} degrees and the angular size of the source 0.005 seconds of arc.

To explain this data, it was assumed that a population difference existed and that maser action was taking place. Pumping processes suggested are infrared radiation and/or chemical processes.

On Dec. 16, 1968, a group at Berkeley[9] announced the detection of NH_3 emission in the 23.7 GHz range, corresponding to J=1, K=1, and J=2, K=2 inversion transitions, from the same dense interstellar cloud that OH absorption was observed. Figure 9(B) gives the pertinent energy levels of NH_3 involved.

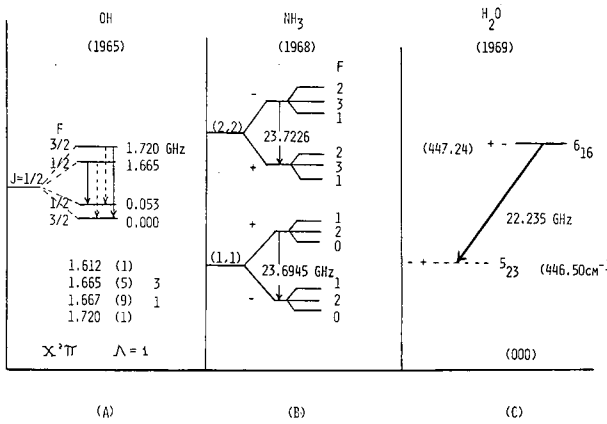

Fig 9 *Molecular lasing sources in the sky*

On Feb. 15, 1969, the same Berkeley[10] group reported detecting emission from the 6_{-5} to 5_{-1} transition of the ground state (000) of H_2O from the direction of SgrB2, the Orion Nebula, and W49. The energy levels associated with this transition are given in Figure 9 (C).

The earth's atmosphere is highly absorbing throughout the submillimeter region where most molecular lasers operate. It is interesting to speculate how many molecular laser signals one could see from a satellite where the absorption would be absent. A molecular laser would be the ideal local oscillator to use in a superheterodyne system since its frequency would just be the Doppler frequency away from any stellar line.

Even on a modest budget, a molecular laser system can be built to achieve hundreds of laser lines scattered through 1- to 1,000-micron range covering the near infrared submillimeter area of the spectrum.

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