Optically Pumped Submillimeter-Wave Sources

T. Y. CHANG, MEMBER, IEEE

Abstract—**Optical pumping of polar molecules by means of infrared lasers is expected to lead to thousands of laser lines throughout the submillimeter-wave region. Already, 282 new laser lines between 34 μm and 1.814 mm in 18 different molecules have been reported. Very stable CW operation, very high pulsed output, linear polarization, high gain, and many other advantages over discharge excitation are offered.**

THE DEVELOPMENT of the submillimeter-wave (SMMW) region has been greatly hampered in the past by the lack of good sources with extensive spectral coverage. This kind of obstacle is now rapidly disappearing due to the development of optically pumped SMMW lasers. Since 1970, 282 new SMMW laser lines produced by optical pumping have been reported. Many of these laser lines provide stable CW output at the milliwatt level and some of them are capable of delivering pulsed output at the multikilowatt level. The technique is very general and should eventually lead to thousands of laser lines throughout the extended SMMW region.

The basic principles of optically pumped SMMW lasers are quite simple. Shown in Fig. 1 is a partial energy-level diagram for a symmetric top molecule. Two vibrational states of the molecule are shown, the lower of which is either the ground vibrational state or a low-lying vibrational state. Each vibrational state consists of a large number of rotational levels with different quantum numbers J and K. In optical pumping, molecules are excited by means of an infrared laser beam from a certain rotational level in the lower vibrational state to a rotational level (J',K') in the upper vibrational state. Since the thermal population in the upper vibrational state is very small, an inversion of population between levels (J',K') and $(J'-1,K')$ is readily established. Laser action can then be obtained on the rotational transition between these levels provided that the molecule has a permanent dipole moment. Sometimes, laser action on the cascade transition $(J'-1,K')$–$(J'-2,K')$ and laser action in the lower vibrational state due to the depletion of population from the starting level are also observed.

Since the photon energy of SMMW radiation is $\lesssim kT$, the energy separation between these rotational levels is quite small. The close spacing of energy levels makes it very difficult to achieve population inversion by other means, e.g., by electrical discharge. This kind of difficulty does not arise in optical pumping due to the high degree of monochromaticity of the pump radiation. However,

Manuscript received May 6, 1974.
The author is with Bell Laboratories, Holmdel, N. J. 07733.

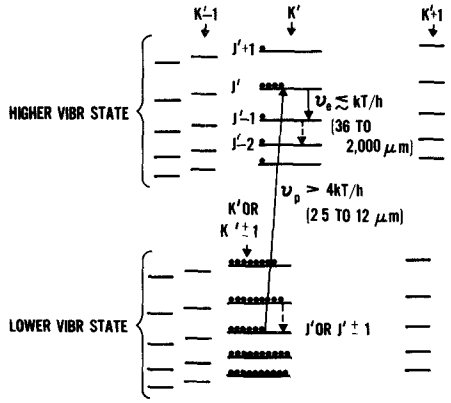

Fig. 1. Partial energy-level diagram for a symmetric top molecule, showing transitions in an optically pumped SMMW laser.

a close coincidence between the pump wavelength and the absorption wavelength is required. Fortunately, there are many infrared laser lines and there are also many absorption lines for each molecule. Consequently, the probability of spectral coincidence is reasonably high.

The operating pressure of optically pumped SMMW lasers is usually between 0.01 and 1 torr. At these pressures the absorbing transition is predominantly Doppler broadened, while the emission transition is mainly collision broadened. In CW operation only molecules of a certain velocity class are excited into the upper laser level by the monochromatic pump radiation.

Two kinds of difficulty are encountered in the operation of these lasers at higher pressures. One is collisional relaxation among rotational levels, which is particularly rapid for polar molecules. The other is self-absorption of the emission frequency by rotational transitions in lower vibrational states, which may be negligible at lower pressures due to slight relative shifts of rotational spectra associated with different vibrational states.

The small-signal gain in an optically pumped SMMW laser is given in MKS units by [1]

$$\gamma = \frac{4\pi}{3h\epsilon_0\lambda_{ij}\Delta\nu}\cdot\mu_{ij}^2\cdot\left(n_i - \frac{g_i}{g_j}n_j\right) \tag{1}$$

where $\Delta\nu$ is the Lorentz linewidth full width at half maximum (FWHM), μ_{ij} is the dipole moment of the $i \rightarrow j$ transition, and g_i and g_j are statistical weights of i and j levels, respectively. For a symmetric top molecule

$$g_i = g_{J'} = 2J' + 1$$

$$g_j = g_{J'-1} = 2J' - 1$$

and [2]

$$\mu_{ij}^2 = \mu_v^2 \frac{J'^2 - K'^2}{J'(2J' + 1)} \tag{2}$$

where μ_v is the permanent dipole moment of the molecule in the excited vibrational state. The value of μ_v is usually on the order of 1 debye (3.3356×10^{-30} C·m), which is very large compared to the dipole moment of, say, the CO_2-laser transition (0.0356 debye). This large value of dipole moment more than offsets the unfavorable factor of having a large value of λ_{ij} in the denominator of (1). The ratio of inversion density over linewidth that can be achieved is on the order of 10^{11}–10^{12} cm^{-3} MHz^{-1}. When these typical values are inserted into (1), we find that the range of small-signal gain that can be expected for a laser of this kind is from 0.01 m^{-1} to 10 m^{-1}, i.e., 1 percent/m–1 percent/mm, which is certainly adequate for achieving laser action.

The inversion density that can be achieved depends to a large extent on the population available at the starting rotational level in the lower vibrational state. Based on this fact, one can show that a given molecule will exhibit the highest gain when the emission frequency is about equal to $(4BkT/h)^{1/2}$, where B is the rotational constant of the molecule. In order for the gain to be within an order of magnitude of this maximum value, the emission frequency should fall within the following bounds:

$$(0.2BkT/h)^{1/2} < \nu_e < (20BkT/h)^{1/2}.$$

For the molecule CH_3F ($B = 0.84$ cm^{-1}), this frequency range turns out to be 6 cm^{-1}–60 cm^{-1}. Other molecules would cover a similar range either somewhat higher or somewhat lower in frequency. Asymmetric top molecules usually cover a larger frequency range. The SMMW region is hence very adequately covered by rotational transitions.

Statements made in the preceding paragraph are qualitatively reflected by the spectral distribution of reported laser lines as shown in Fig. 2. The wavelengths span a total of six octave bands from approximately 30 μm to 2000 μm, and the distribution is peaked near the center of the SMMW region. Notice that the average spacing between reported laser lines is already only a fraction of cm^{-1} in some octave bands as shown in the lower half of Fig. 2.

A representative experimental system is shown schematically in Fig. 3. The principal device in the system is a meter-long internal-mirror hole-coupled SMMW laser resonator which is filled with the molecules to be excited to a typical pressure of 0.1 torr. The SMMW output emanating from the cone-shaped coupling hole is first deflected by a beam-splitter flange and then exits from the vacuum vessel as a collimated beam through a polyethylene lens/window. The infrared pump beam is focused into the resonator through a small window on the beam-splitter flange and through the coupling hole on the output mirror. The pump beam is then reflected many times between the resonator mirrors until it is completely absorbed by the gas.

The infrared pump laser, whether of the CW variety

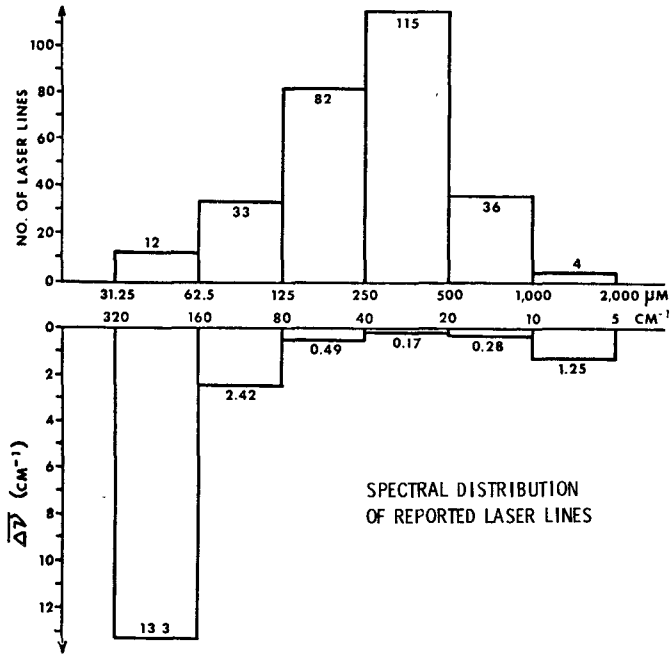

Fig. 2. Spectral distribution of reported laser lines. The extended SMMW region is divided into six octave bands. The average spacing between laser lines in each octave band is shown in the lower half of the figure.

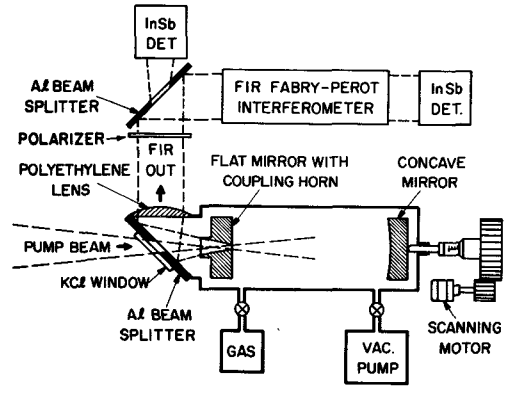

Fig. 3. Schematic diagram of a representative optically pumped SMMW laser system.

or of the high-power pulsed TEA variety, is usually equipped with a diffraction-grating end mirror to permit operation on one of many transitions available from the laser.

For the detection of the SMMW signal, the following liquid-helium-cooled detectors with submicrosecond response are often used: Ge:Ga photoconductor for $\lambda < 170$ μm, InSb hot-electron bolometer for $\lambda > 150$ μm, and GaAs photoconductor for 80 μm $< \lambda <$ 500 μm. For CW operation, one might find it more convenient to use one of the slower room-temperature thermal detectors such as Golay cell, vacuum thermocouple, or flake thermistor. These room-temperature detectors are less sensitive but still have NEP's lower than 10^{-9}-W/Hz$^{1/2}$ bandwidth. The polarization of the SMMW output is examined by placing a far-infrared grid polarizer in front of the detector.

The wavelength of the SMMW laser output can be estimated with about 1-percent accuracy by using the laser resonator itself as a scanning interferometer. The use of an external far-infrared Fabry–Perot interferometer (which uses fine metal-mesh mirrors) as indicated in Fig. 3 can increase the accuracy to about one part in 10^4. A still higher accuracy of one part in 10^6 or better can be achieved in frequency measurement by beating the SMMW signal with the harmonics of an accurately monitored tunable microwave signal in a point-contact diode.

In designing a resonator of the type shown in Fig. 3, the mirror diameters must be made sufficiently large to avoid excessive diffraction losses. The Fresnel number of the resonator should be greater than 1.5 at the longest wavelength of interest. The curvature of the mirrors should also be chosen with care. For instance, the pump beam may not be efficiently absorbed in a confocal or a semiconfocal resonator, because the pump beam is refocused onto the coupling hole and reflected out of the resonator after only a few bounces in the resonator. Shown in Fig. 4 is an experimental curve showing the relative intensity of pump radiation that is reflected from a weakly absorbing resonator as a function of the ratio between the resonator length and the radius of curvature of the concave mirror (the output mirror being flat). It is seen that the L/R ratio should be between 0.3 and 0.4 or between 0.6 and 0.7 for maximum pump absorption.

Metal-waveguide resonators offer an attractive alternative to open resonators for compactness [3]. Shown in Fig. 5 is a metal-waveguide resonator constructed from a 23-mm-bore gold-coated brass tubing which is 80 cm long and is terminated at two ends by adjustable plungers with coupling holes. Also shown in the background is a dielectric-waveguide CO_2 pump laser which is also very compact.

The first optically pumped SMMW laser was a CH_3F laser [4] with six laser transitions, as indicated in a partial energy-level diagram in Fig. 6. The pump source was a Q-switched CO_2 laser operating on the $P(20)$ line of the 9.6-μm band. Its spectral coincidence with the $Q(12)$ transition of the ν_3 vibrational band of CH_3F was previously

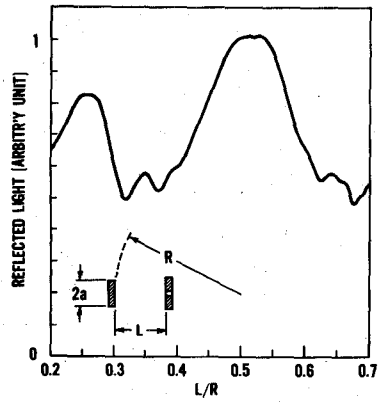

Fig. 4. Relative intensity of the pump radiation reflected from a weakly absorbing resonator as a function of the ratio between the resonator length L and the radius of curvature R of the concave mirror.

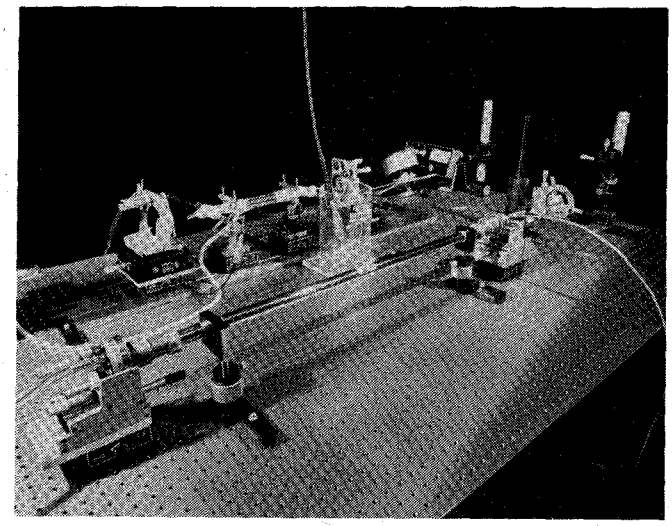

Fig. 5. A compact SMMW laser system comprising a metal-waveguide SMMW resonator and a dielectric-waveguide CO_2 laser (shown in background). (Courtesy of D. T. Hodges.)

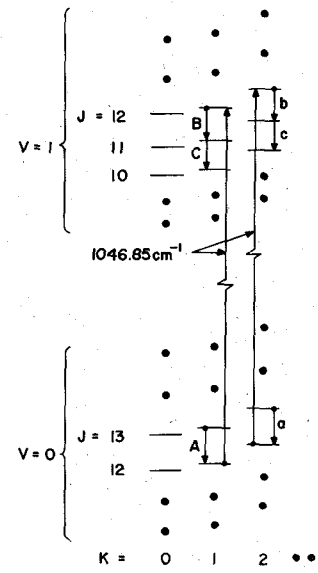

Fig. 6. Partial energy-level diagram for CH_3F showing pump and SMMW laser transitions.

identified in a study of the molecule as a saturable absorber for CO_2 lasers [5]. Two K components, $Q(12,1)$ and $Q(12,2)$, of the absorbing transition were found to be accessible to the CO_2-laser line. As a result, the emission lines also consisted of two K components separated by 30–40 MHz. The output of the laser as a function of the resonator length is shown in Fig. 7, showing periodic resonance peaks associated with the six different transitions. The identification of these laser transitions was ascertained by comparing accurately measured frequencies of A, a (452-μm) transitions with the known spectroscopic data of the ground vibrational state. Accurate frequency measurements of B, b (496-μm) and C, c (541-μm) transitions, on the other hand, produced new accurate spectroscopic data for the ν_3 vibrational state of CH_3F.

One interesting observation in this and other optically

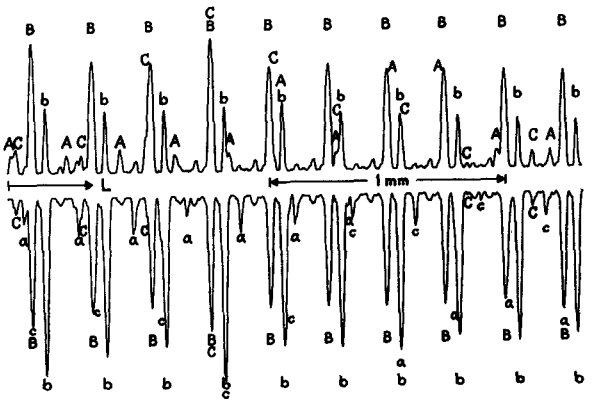

Fig. 7. Output of a CH_3F laser as a function of the resonator length showing periodic resonances corresponding to different laser transitions. The lower trace is obtained at an approximately 50-MHz higher pump frequency.

pumped SMMW lasers is that when the pump beam is linearly polarized, the SMMW output is also almost always linearly polarized. The plane of polarization has a definite orientation for each SMMW transition and is either parallel or perpendicular to the plane of polarization of the input beam. This phenomenon is due to the fact that the dipole moment of any transition, whether vibrational–rotational or pure rotational, is oriented either perpendicular or parallel to the space-fixed total-angular-momentum vector of the molecule. Molecules that happen to have their total-angular-momentum vector oriented relative to the pump field in the direction for maximum pump absorption therefore absorb and emit more strongly than other molecules with other orientations. As a result, the excited medium exhibits a polarized gain pattern and the laser oscillates with its electric field polarized in the preferred direction. The case for a symmetric top molecule is illustrated in Fig. 8. Linear polarization of the SMMW output is not only of great practical value but also provides spectroscopic clues to the type of transition involved in the pumping process.

Between 1970 and 1973, a total of 282 optically pumped SMMW laser lines in 18 different molecules, ranging in wavelength from 34 to 1814 μm, have been reported. A

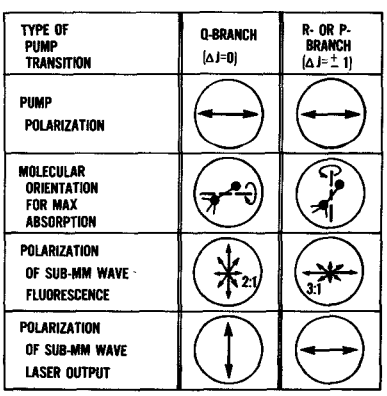

Fig. 8. Dependence of the polarization of SMMW output on the type of pump transition, showing the preferred orientations of the total angular momentum, polarization patterns of spontaneous SMMW radiation, and the polarization planes of the SMMW laser.

summary organized according to the molecular type is given in Table I. Some additional data that are yet to be published by this author are also included in parentheses in the table. Except for the HF molecule which was pumped by a TEA HF laser [6] and two lines in NH_3 [7] which were pumped by an N_2O laser, all of the laser lines were excited by a CO_2 laser.

A short list of 39 strong CW laser lines pumped by a CO_2 laser is given in Table II. An output power of roughly 1 mW or more can be expected for each of these laser lines.

In CW operation a spectral coincidence of better than 100 MHz between the pump frequency and the absorption line is usually required. This requirement is considerably relaxed by the use of a high-power TEA laser. In fact, laser action has been observed by pumping an absorbing transition as much as 40 collisional widths off resonance [10]. It has also been demonstrated that by applying a strong electric field, a normally forbidden transition can be made active and brought into spectral coincidence with a pump source to produce SMMW laser emission [16].

The use of high-power TEA lasers has lead to the observation of SMMW superradiant emission in HF [6], [17] and CH_3F [18]–[20]. In HF, both the difference between forward and backward gains [6] and the evolution of the superradiant pulse [17] have been studied experimentally and compared to theories. In CH_3F, superradiant pulses with output power as high as 30 kW [20] have been reported for the 496-μm line.

Not all the molecules listed in Table I have been investigated thoroughly. Nevertheless, it is apparent that by using a CO_2 laser, one can find on the average one dozen or more SMMW laser lines in each molecule, and three dozen or more lines in a molecule with OH group. There are more than a hundred other volatile molecules with absorption bands in the 10-μm range. One can therefore expect more than a thousand SMMW laser lines to result from a thorough investigation of these molecules with a CO_2 laser. The use of N_2O, isotopic CO_2, CO, HF, and other infrared lasers should further increase the number of SMMW laser lines by severalfold. Finally, an enormous number of SMMW laser lines should result from the use of tunable infrared sources such as spin-flip Raman lasers and parametric oscillators.

For applications in magnetospectroscopy, the optically pumped SMMW sources have proved to be superior to discharge-excited HCN and H_2O lasers, particularly in their large selection of wavelengths, their high-power capabilities for nonlinear studies, and their high stability (when excited by an ultrastable CO_2 laser) for high resolution experiments [21], [22]. The very high-power capability of SMMW lasers is also of interest for use in gas breakdown and plasma heating under cyclotron resonance conditions [23]. Optically pumped SMMW lasers should also find applications in other aspects of materials research, plasma diagnostics, imaging and probing, metrology, ellipsometry, ranging, radiometry, astronomy, and communication.

In addition to producing useful SMMW sources, the optical pumping process also provides a new form of

TABLE I
NUMBER OF OPTICALLY PUMPED SMMW LASER LINES PRODUCED IN VARIOUS MOLECULES

Type of Molecule	Mol. Formula	No. of Lines	Ref.
Diatomic Molecule	HF	6	6
Symmetric Top	CH_3F	16	4,8,9,10
W/O Internal	CH_3Cl	4 + (20)	10
Rotation	CH_3Br	(35)	
	CH_3I	(20)	
	CH_3CN	7 + (24)	9
	CH_3CCH	8 + (7)	9
Symm. Top. W. Inversion	NH_3	30	7,10,12,13,16
Symm. Top. W. Internal Rot.	CH_3CF_3	1 + (27)	10,
Asymmetric Top	CH_2CHCl	3	8
W/O Internal Rotation	CH_2CHCN	5	11,
	CH_2CF_2	13	11,15
	O_3	3	12,
Asymmetric Top	CH_3OH	41	8,10,12,15
W. OH Group	HCOOH	43	12,14
	CH_2OHCH_2OH	42	14.
Asymm. Top W. NH_2 Group	CH_3NH_2	32	11,14,
Asymmetric Top	CH_3OCH_3	6	14.
W. Heavier	CH_3CH_2F	18	12.
Internal Rotation	CH_3CHF_2	4	10,15
Group			
TOTAL 18 + (2) Molecules		282 + (133)	

Note: Some data yet to be published by the author are included in parentheses.

TABLE II
STRONG CW LASER LINES PUMPED BY A CO_2 LASER

λ (μm)	MOLECULE	λ (μm)	MOLECULE	λ (μm)	MOLECULE
37.5	CH_3OH	251.9	CH_3F	496.1	CH_3F
40.2	CH_3OH	277	CH_2OHCH_2OH	512	HCOOH
41.7	CH_3OH	369.1	CH_3OH	520	CH_3OCH_3
70.1	CH_2OHCH_2OH	372.7	CH_3F	533	CH_3CHF_2
70.6	CH_3OH	372.9	CH_3CN	554	CH_2CF_2
95.8	CH_2OHCH_2OH	388	HCOOH	568	CH_2CF_2
117.1	CH_2OHCH_2OH	392.3	CH_3OH	570.5	CH_3OH
118.8	CH_3OH	393.6	HCOOH	647.9	CH_3CCH
118.9	CH_2OHCH_2OH	414	HCOOH	713.7	CH_3CN
147	CH_3NH_2	415	CH_2CF_2	884	CH_2CF_2
170.6	CH_3OH	428	HCOOH	1174.9	CH_3CCH
192.8	CH_3F	458	CH_3CHF_2	1221.8	$C^{13}H_3F$
197	CH_2OHCH_2OH	492	CH_3OCH_3	1814.4	CH_3CN

precision emission spectroscopy, particularly for the excited vibrational states of molecules. New spectroscopic data can be obtained sometimes by precise measurements of the emission frequencies [4] and sometimes from correlations among many emission and absorption wavelengths associated with the same molecule [9].

The advantages of optically pumped SMMW lasers are manyfold. 1) CW output power at the milliwatt level and pulsed output power at the kilowatt level are available at a large number of stable discrete wavelengths covering the entire extended SMMW region. 2) Pump sources, such as the CO_2 laser, are well developed and highly stable. 3) Discharge-associated fluctuations, thermal drift, and molecular dissociation are completely absent in the SMMW resonator. 4) The output is linearly polarized without need for any polarizing element. 5) The overall efficiency is good because the pump lasers are efficient and the pump energy is deposited only in the intended energy level. 6) A very long resonator is not required because the gain is reasonably high. In some cases, the gain is sufficiently high to permit generation of high-power superradiant pulses in a traveling-wave configuration. 7) It is feasible to construct a compact and perhaps permanently sealed system.

With so many outstanding features, the optically pumped SMMW lasers will undoubtedly contribute immensely to the further development of the SMMW region.

REFERENCES

[1] J. P. Gordon, "Elements of laser theory," in *Laser Technology and Applications*, S. L. Marshall, Ed. New York: McGraw-Hill, 1968.

[2] C. H. Townes and A. L. Schawlow, *Microwave Spectroscopy*. New York: McGraw-Hill, 1955.

[3] D. T. Hodges and T. S. Hartwick, "Waveguide laser for the far infrared (FIR) pumped by a CO_2 laser," *Appl. Phys. Lett.*, vol. 23, pp. 252–253, Sept. 1, 1973.

[4] a) T. Y. Chang and T. J. Bridges, "Submillimeter wave laser action in optically pumped CH_3F," in *Proc. Symp. Submillimeter Waves*, J. Fox, Ed. Brooklyn, N. Y.: Polytechnic Press, 1970, pp. 93–98.

b) ——, "Laser action at 452, 496, and 541 μm in optically

pumped CH_3F," *Opt. Commun.*, vol. 1, pp. 423–426, Apr. 1970.
[5] T. Y. Chang, C. H. Wang, and P. K. Cheo, "Passive Q-switching of the CO_2 laser by CH_3F and PF_5 gases," *Appl. Phys. Lett.*, vol. 15, pp. 157–159, Sept. 15, 1969.
[6] N. Skribanowitz, I. P. Herman, R. M. Osgood, Jr., M. S. Feld, and A. Javan, "Anisotropic ultrahigh gain emission observed in rotational transitions in optically pumped HF gas," *Appl. Phys. Lett.*, vol. 20, pp. 428–431, June 1, 1972.
[7] T. Y. Chang, T. J. Bridges, and E. G. Burkhardt, "Cw laser action at 81.5 and 263.4 μm in optically pumped ammonia gas," *Appl. Phys. Lett.*, vol. 17, pp. 357–358, Nov. 1, 1970.
[8] ——, "Cw submillimeter wave laser action in optically pumped methyl fluoride, methyl alcohol, and vinyl chloride gases," *Appl. Phys. Lett.*, vol. 17, pp. 249–251, Sept. 15, 1970.
[9] T. Y. Chang and J. D. McGee, "Millimeter and submillimeter wave laser action in symmetric top molecules optically pumped via parallel absorption bands," *Appl. Phys. Lett.*, vol. 19, pp. 103–105, Aug. 15, 1971.
[10] H. R. Fetterman, H. R. Schlossberg, and J. Waldman, "Submillimeter lasers optically pumped off resonance," *Opt. Commun.*, vol. 6, pp. 156–159, Oct. 1972.
[11] S. F. Dyubko, V. A. Svich, and L. D. Fesenko, "Submillimeter-band gas laser pumped by a CO_2 laser," *Pisma Zh. Eksp. Teor. Fiz.*, vol. 16, pp. 592–594, Dec. 5, 1972 (transl.: *JETP Lett.*, vol. 16, pp. 418–419).
[12] R. J. Wagner, A. J. Zelano, and L. H. Ngai, "New submillimeter laser lines in optically pumped gas molecules." *Opt. Commun.*, vol. 8, pp. 46–47, May 1973.
[13] K. Gullberg, B. Hartmann, and B. Kleman, "Submillimeter emission from optically pumped $^{14}NH_3$," *Physica Scripta*, vol. 18, pp. 177–182, 1973.
[14] T. K. Plant, P. D. Coleman, and T. A. DeTemple, "New optically pumped far-infrared lasers," *IEEE J. Quantum Electron.* (Notes and Lines), vol. QE-9, pp. 962–963, Sept. 1973.
[15] D. T. Hodges, R. D. Reel, and D. H. Barker, "Low-threshold CW submillimeter- and millimeter-wave laser action in CO_2-laser-pumped $C_2H_4F_2$, $C_2H_2F_2$, and CH_3OH," *IEEE J. Quantum Electron.* (Notes and Lines), vol. QE-9, pp. 1159–1160, Dec. 1973.
[16] H. R. Fetterman, H. R. Schlossberg, and C. D. Parker, "Cw submillimeter laser generation in optically pumped Stark-tuned NH_3," *Appl. Phys. Lett.*, vol. 23, pp. 684–686, Dec. 15, 1973.
[17] N. Skribanowitz, I. P. Herman, J. C. McGillivray, and M. S. Feld, "Observation of Dicke superradiance in optically pumped HF gas," *Phys. Rev. Lett.*, vol. 30, pp. 309–312, Feb. 19, 1973.
[18] F. Brown, E. Silver, C. E. Chase, K. J. Button, and B. Lax, "10-W methyl fluoride laser at 496 μm," *IEEE J. Quantum Electron.* (Corresp.), vol. QE-8, pp. 499–500, June 1972.
[19] T. A. DeTemple, T. K. Plant, and P. D. Coleman, "Intense superradiant emission at 496 μm from optically pumped methyl fluoride," *Appl. Phys. Lett.*, vol. 22, pp. 644–646, June 15, 1973.
[20] F. Brown, S. R. Horman, A. Palevsky, and K. J. Button, "Characteristics of a 30-kW-peak, 496 μm, methyl fluoride laser," *Opt. Commun.*, vol. 9, pp. 28–30, Sept. 1973.
[21] H. R. Fetterman, H. R. Schlossberg, and J. Waldman, "Submillimeter spectroscopy," *Laser Focus*, vol. 8, pp. 42–44, Sept. 1972.
[22] H. R. Fetterman, J. Waldman, C. M. Wolfe, G. E. Stillman, and C. D. Parker, "Identification of donor species in high-purity GaAs using optically pumped submillimeter lasers," *Appl. Phys. Lett.*, vol. 21, pp. 434–436, Nov. 1972.
[23] B. Lax and D. R. Cohn, "Cyclotron resonance breakdown with submillimeter lasers," *Appl. Phys. Lett.*, vol. 23, pp. 363–364, Oct. 1, 1973.

High Power Optically Pumped Far Infrared Lasers

THOMAS K. PLANT, STUDENT MEMBER, IEEE, LEON A. NEWMAN, STUDENT MEMBER, IEEE, EDWARD J. DANIELEWICZ, STUDENT MEMBER, IEEE, THOMAS A. DeTEMPLE, MEMBER, IEEE, AND PAUL D. COLEMAN, FELLOW, IEEE

Abstract—**Intense superradiant laser action in the far infrared (FIR) has been observed in several gases optically pumped with a CO_2 transversely excited atmospheric-pressure (TEA) laser. A maximum FIR power of 100 kW was observed from CH_3F at 496 μm. Characteristics of the system and possibilities of scaling to higher powers are also discussed.**

I. INTRODUCTION

SELECTIVE excitation of a pure rotational lasing transition by optically pumping with another laser was first reported by Chang in 1970 [1]. In this instance a Q-switched CO_2 laser operating on the P(20) 9.6-μm transition was used to pump the $v = 1$, $J = 12$ level of CH_3F giving a 496-μm pure rotational lasing transition to $v = 1$, $J = 11$. Since that time, many similar lasing transitions have been reported in various molecules pumped with CO_2, HF, and N_2O lasers. These optically pumped lasers require optical cavities and produce output pulses typically < 100 W. Recently, intense (~1-kW) superradiant laser action has been reported on the 496-μm pure rotational transition in optically pumped CH_3F [2], [3]. It is the purpose of this paper to present a study of similar intense transitions observed in other molecules and in CH_3F.

II. EXPERIMENT

In Fig. 1 is shown a diagram of the experiment. The grating-tuned transversely excited atmospheric-pressure (TEA) CO_2 laser was of the parallel electrode preionization type and was capable of producing megawatt (MW) pulses throughout the 9.6- and 10.6-μm bands. Provision was also made for suppressing the normal self-mode-locked characteristic of CO_2 and forcing the laser to oscillate on a single longitudinal mode near line center [4]. The far infrared (FIR) cell was 40-mm-ID pyrex tubing,

Manuscript received May 14, 1974. This work was supported by the Air Force Office of Scientific Research, the National Science Foundation, and the University of Illinois Industrial Affiliates Program.

The authors are with the Electro-Physics Laboratory, Department of Electrical Engineering, University of Illinois, Urbana, Ill. 61801.