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Mid-Infrared Optically Pumped Lasers

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Mid-Infrared Optically Pumped Lasers

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MID-INFRARED OPTICALLY PUMPED LASERS

BY

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ABSTRACT

Optically pumped lasers emitting in the middle-infrared spectral region are reviewed. General features of these lasers are discussed, and published data on them are summarized. Approximate lasing wavelengths are indicated, and a few of the more important mid-infrared optically pumped lasers are treated in more detail.

I. INTRODUCTION

Over the past several years optically pumped lasers have become a major part of laser technology. Although far-infrared (FIR) sources have dominated this field since 1970, developments at shorter wavelengths during recent years have made mid-infrared (MIR) and visible-near-infrared lasers important additions to the community.

In this report we review the status of mid-infrared lasers that are optically pumped by coherent sources. The latter sources can be the standard higher energy lasers HF, DF, CO, CO_2, and others. The composite wavelength range covered by these conventional infrared lasers is 2.6 to 11 μm, but the coverage is in a piecemeal fashion, as we will illustrate later. Optically pumped lasers offer the possibility of sources that fill these wavelength gaps and significantly extend the useful range of laser frequencies.

Sources exist that can, in principle, tune continuously across the entire MIR spectrum. Semiconductor laser diodes represent one example, but because of their low power are useful only in high-resolution spectroscopy. Higher power, continuously tunable lasers based on nonlinear processes, e.g., optical parametric oscillators, are well known but these are characterized by limited power, operating difficulties, and high cost. Mid-infrared optically pumped lasers with their capability of high powers can play a useful role in applications requiring significant spectral irradiance. Those disciplines expected to use these lasers are photochemistry, photophysics, isotope enrichment, and atmospheric species monitoring.

First, we will briefly describe general features of these lasers and summarize the numerous MIR optically pumped lasers now available. Several of the more interesting examples will be emphasized. Later, optical configurations used for these lasers will be discussed, and finally, some of the important considerations regarding the interaction of the pump laser with the pumped medium will be mentioned.

II. GENERAL FEATURES

Excitation of the lasing medium is accomplished by directing the output radiation from the pump laser into a cavity containing the medium. As shown in Fig. 1, the cavity consists of a cell containing the gas (all MIR lasers reported to date have been gas phase) and an optical resonator. If the pump frequency is resonant or near-resonant with a vibrational-rotational transition of the pumped molecule, lasing can occur at a wavelength longer than that of the pump. Unlike incoherent source optical pumping essentially all the pump laser output can be effective in creating the inversion.

Whereas far-infrared (or submillimeter) lasers operate on pure rotational transitions, MIR lasing occurs on vibrational-rotational (VJ) transitions. Shown in Fig. 2 are several energy level schemes first pointed out by Letokhov,[1] which sketch the physics of these lasers. The first, type A, is one in which a combination level of the molecule is excited and lasing occurs on a hot band. Because the pump transition involves two or more vibrational quanta, absorption of the pump beam is often small, but gain on the laser transition can be large since a single-quantum transition occurs. A special case of type A is the excitation of an overtone transition in a diatomic or a polyatomic molecule.

In the second case (B) a fundamental level is pumped and emission occurs within a difference band to a lower level. Here, the pump absorption coefficient can be large, but since the lasing transition demands a change of two or more quanta, optical gain will often be low. Finally, in type C the pump and lasing can occur on different lines within the same fundamental band. For example, excitation of an R-branch line can be followed by lasing on a P-branch line. The cross section for both excitation and emission can be large in this case.

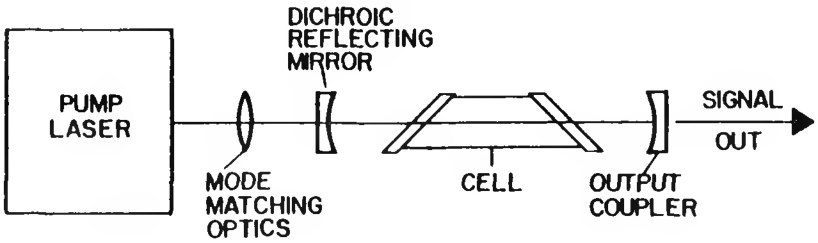

Fig. 1.
Sketch of mid-infrared optically pumped laser apparatus. Several variations have been used.

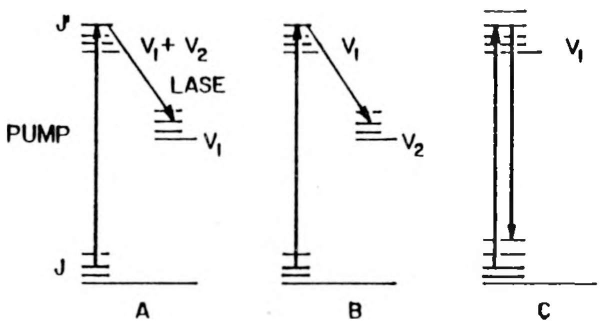

Fig. 2.
Molecular energy level schemes describing mid-infrared optically pumped lasers. In (A) a combination level is excited and lasing is on a hot band. In (B) a fundamental mode is pumped and emission is on a difference band. In (C) both pump and lasing occur in the same band.

In each of these cases lasing will most frequently occur from the vibrational-rotational level excited by the pump, and in most cases only a single level in the molecule will be optically pumped. Collisions, however, can induce lasing from neighboring rotational levels or even from other vibrational levels. Also, cascade lasing can occur, i.e., stimulated emission on one transition can create sufficient inversion on a connected lower transition to induce lasing thereon. These mechanisms are illustrated in Fig. 3.

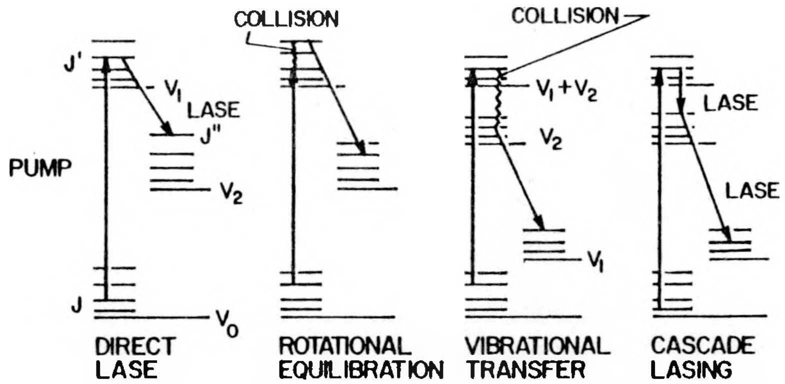

Fig. 3.
Various mechanisms for exciting the upper laser level in mid-infrared optically pumped lasers. In the first case, this level is populated directly by the pump. In the second case, rotational equilibration occurs subsequent to the pump pump step, often leading to many different laser lines. In the third case, V-V collisions occurring after the pump step can effect lasing from a vibrational level different from the one pumped. In the fourth diagram, lasing on an upper transition creates an inversion on a connected lower transition that lases.

To the list of direct optical pumping schemes must be added the technique of transfer optical pumping in which the pumped molecule transfers by collision (V-V most frequently) its excitation to the lasing species. It appears that all reported lasing due to transfer optical pumping has occurred on difference bands. All the aforementioned optical pumping schemes are well represented in the literature.

Each of the spectroscopic types of optically pumped laser has advantages and disadvantages from the standpoint of performance. Type A has a somewhat predictable limitation in pressure scaling for two reasons: First, V-V collisional break-up of the excited combination level can be fast, and second, there is automatically a ground-state absorption band at nearly the same frequency as the lasing band. Collision-broadening of this absorption will ensure a significant cavity loss at some pressure. High-pressure or condensed-phase lasers based upon a type-A level scheme are not as likely as for type B.

Because they often have a small gain coefficient on the lasing transition, type-B lasers can be difficult to operate under normal conditions. However, in many cases systems of this type are scalable with pressure because long collisional relaxation times are often associated with fundamental molecular modes, and because there is normally no ground state absorption near the lasing wavelength.

It was pointed out above that in all cases of transfer optical pumping, the lasing molecule operated in the type-B mode. This is because of the slower rate associated with collisional pumping than with direct optical pumping, and the consequent requirement for the lasing level to be capable of withstanding collisions before lasing.

Type-C lasers can be very efficient because of large cross sections associated with both the pump and lasing steps. Here one can expect stimulated Raman effects to contribute to the emission in many cases.

III. INDIVIDUAL SYSTEMS

As of this writing 26 different molecules have lased in the MIR when pumped by 6 types of pump laser. We will not attempt to enumerate the many lasing wavelengths, which lie between 2.6 and 35 μm. In Table I salient information on MIR optically pumped lasers is summarized. We will briefly discuss many of these optically pumped lasers and partition the discussion according to pump laser as we did in the Table.

HF Pump Laser

This laser emits up to 10-MW pulses as a convenient laboratory tool. The wavelength range is 2.6 to 2.9 μm, consisting of approximately 25 distinct frequencies separated by $\sim 40\ cm^{-1}$. Normally lasing on the 1→0, 2→1, and 3→2 vibrational transitions, this laser prefers the multiline mode and will not efficiently operate single line. This feature makes the HF laser convenient for optical pumping surveying research but unhappily is largely responsible for the inefficiency of this source in specific cases, since usually only a single line is effective in exciting a particular molecule. The short wavelength of the HF laser allows the generation of optically pumped wavelengths considerably shorter than those available by CO_2 laser pumping and therefore nicely augments this latter source.

Any molecule pumped by the HF laser must be excited to a combination level (except for hydrides) since the lasing frequencies are in the 3500 cm^{-1} range. The first laser pumped by the HF laser was, in fact, a hydride, HF gas, the second reported MIR optically pumped laser.[2]

All other HF laser-pumped systems were reported by Los Alamos Scientific Laboratory (LASL), where 10 different molecules have been observed to lase.[3-6] The wavelength range covered was 4.4 to 18 μm. As seen in Table I, isotopic CO_2 and N_2O, $^{15}NH_3$, HCOOH, and $^{13}CS_2$ have been successfully used. In the case of $^{12}C^{18}O_2$ a photon conversion efficiency to 4.3-μm lasing of 20% was measured.[4] This is one of the more interesting of the HF laser-pumped species because four pumping mechanisms are operative: (1) 4.3- and 17-μm hot-band lasing originating on a directly pumped combination level, (2) 17-μm lines cascading from intense 4.3-μm lasing, (3) 4.3-μm lines induced by rotational relaxation within the pumped combination level, and (4) 10-μm lasing subsequent to V-V collisional break-up of the excited combination level (see Fig. 3). High gains associated with hot-band lasing systems were manifested in $^{15}NH_3$ and isotopic CO_2 and N_2O by mirrorless lasing on certain transitions.

Although an obvious extension of HF laser excitation, the use of a DF laser in optical pumping has not been widespread. In the only report of a cw optically pumped MIR laser, Wang et al.[7] observed continuous wave lasing at 10.6 μm from a DF-CO_2 mixture pumped by a combustion-driven DF chemical laser.

HBr Pump Laser

Despite its modest energy output, the HBr laser has been a very important pump source in this field. The first MIR optically pumped laser,[8] reported by Chang and Wood in 1972, used the HBr laser to directly pump the 001 level of CO_2. Under optimal conditions, power in the pump line at 4.2 μm appeared in the 10.6-μm lasing with an efficiency of 40%, the highest reported efficiency for any optically pumped laser.

These same authors soon extended their work to high-pressure operation of CO_2, N_2O, and CO_2/N_2O mixtures.[9-11,13] Lasing at pressures up to 42 atm has been achieved, and completely continuous tuning over 5 cm^{-1} from a higher pressure mixture was reported.[13] It should be emphasized that such high-pressure operation is not generally achievable, and that in this case it was possible because of the type-B energy level scheme and favorable kinetics for these N_2O/CO_2 lasers.

TABLE Ia

CHARACTERISTICS OF OPTICALLY PUMPED LASERS

PUMP LASER	PUMP LINE(s)	LASING MEDIUM	LASING BAND	LASING λ-μm	NUMBER OF λ's	LASING ENERGY, POWER, EFFICIENCY	REFERENCE
	R_1(1-4)	HF	P_1(3-6)	2.6	4		2
	P_1	CO_2(HF)	001–100	10.6	many		3
	$P_2$6	$^{12}C^{18}O_2$	021–020	4.4	"	5 mJ	3,4
			020–010	18	"		3
			021–011	18	"		3
			001–100	10.6	"		3
	$P_2$5	$^{12}C^{16}O^{18}O$	021–020	4.3	"		3
			020–010	17	"		3
			021–011	17	"		3
HF			000–100	10.6	"		3
	$P_2$6	$^{13}C^{16}O^{18}O$	021–020	4.6	"		3,4
			021–011	18	"	1 mJ	3,4
	$P_2$6	$^{15}NH_3$	0201–0101?	14–18	few	0.1 mJ	5
	$P_2$5	HCOOH		5.7	"		6
		$^{15}N_2O$	101–100	4.6	"		6
		$^{14}N^{15}NO$	101–100	4.6	"		6
		$^{15}N^{14}NO$	101–100	4.6	"		6
		$^{13}CS_2$	301–300?	6.9	1		6
DF	many	CO_2(DF)	001–100	10.6		1.5 W	7

TABLE Ib

CHARACTERISTICS OF OPTICALLY PUMPED LASERS

PUMP LASER	PUMP LINE(s)	LASING MEDIUM	LASING BAND	LASING λ-μm	NUMBER OF λ's	LASING ENERGY, POWER, EFFICIENCY	REFERENCE
HBr	P_26	CO_2	001–100	10.6	many	40%	8
	P_38	N_2O	001–100	10.8	"		9
	many	CO_2	001–100	10.3,10.6	"	1 kW	10
	"	$N_2O(CO_2)$	001–100	10.5,10.8	"	600 W	11
	"	$CO_2(HBr)$	001–100	10.6	few		12
			100–010	14	"		12
			001–020	9.6	"		12
			020–010	16	"		12
	many	$N_2O(CO_2)$	001–100	10.46–10.54	continuous	150 W	13
CO	few	COF_2	0200–0100	10			14
CO	many	$O^{13}CS$	001–100	8.6	4	1 mJ	14
DOUBLED CO_2	P(26-34)*	OCS	001–100	8.3	few	5 mJ, 19%	15,16
	P24*	OCS(CO)	001–100	8.3	80	1 mJ, 8%	15,16
	P24*	$CO_2(CO)$	001–100	10.6	many	13 mJ, 34%	15,16
	P24*	$N_2O(CO)$	001–100	10.8		0.4 mJ, 6%	15,16
	P24*	$C_2H_2(CO)$	$\nu_2-\nu_5$	8	4	0.1 mJ, 4%	15,16
	P24*	$CS_2(CO)$	001–100	11.5	11	0.03 mJ, 0.5%	15,16
	P24*	$SiH_4(CO)$		8	6	0.03 mJ, 0.6%	16
	P24*	$O^{13}CS(CO)$	001–100	8.6		3.8%	16
	R30*	CS_2	101–100	6.6	1	0.03 mJ, 0.1%	16

TABLE I c

CHARACTERISTICS OF OPTICALLY PUMPED LASERS

PUMP LASER	PUMP LINE(s)	LASING MEDIUM	LASING BAND	LASING λ-μm	NUMBER OF λ's	LASING ENERGY, POWER, EFFICIENCY	REFERENCE
CO_2	9P20	CO_2	001–100	10.6		14%	17
	9P22	OCS	020–010	19	2		18
	10P12,P14#	SF_6		16	1		19
	9R16	NH_3	ν_2-0	12.8	1	5 kW, 0.3%	20
	9P22	OCS	020–010	19	1	1 kW	21
	few	NH_3	ν_2-0	11.5–12.8	5		22
	2	C_2H_4		10–11	4		22
	9R30	NH_3	ν_2-0	9.3–13.8	40		23
	9P18,P34#	NH_3	$2\nu_2-\nu_2$	6–35	10		24
	9R16	NH_3	ν_2-0	12.8	1	200 kW, 4%	25
	9R12	CF_4	$\nu_2+\nu_4-\nu_2$	16	1	4 mJ, 3%	26
	10P34	NOCℓ	011–001	16	2	3 mJ, 10%	26
	9R16	NH_3	ν_2-0	12.8	1	1 MW, 20%	27
	9R16	NH_3	ν_2-0	12.8	1	5 kW, 9%	28
	many	CF_4	$\nu_2+\nu_4-\nu_2$	16	12	4 mJ, 3%	29
	3	CF_3I	$\nu_2+\nu_3-\nu_3$	14	3		29
	many	NH_3	ν_2-0	11–13	8	35 mJ, 12%	29
	"	NOCℓ	011–001	16	10	3 mJ, 10%	29
	"	$^{12}CF_4$	$\nu_2+\nu_4-\nu_2$	16	28	25 mJ	30
	"	$^{13}CF_4$	$\nu_2+\nu_4-\nu_2$	16	31	few mJ	30
	"	$^{14}CF_4$	$\nu_2+\nu_4-\nu_2$	16	25	"	30

A blank entry indicates relevant data not available or were very uncertain.
Where two molecules are listed, the one in parentheses is pumped and transfers to the other, which lases. A* designates CO_2 lines, which are doubled for pumping.
A # indicates that two different pump laser lines were necessary to achieve lasing.

Also using the HBr laser, Osgood[12] pumped HBr-CO_2 mixtures, and the resulting V-V transfer to CO_2 produced 9.6 or 10.6 μm lasing. This lasing, in turn, cascade-pumped lower transitions to produce 16- and 14-μm lasing, respectively.

CO Pump Laser

Pulsed CO lasers on the laboratory scale can deliver energies of a few joules, but the peak optical power is severely reduced by long pulse lengths, usually 10 to 100 μs in duration. The other reason for the low spectral brightness of this source is the multiline (>20 lines) nature of the output, which cannot be efficiently converted to single line. Because of these problems this laser appears to be a poor pump selection for use with optically pumped lasers. However, Nelson et al.[14] have succeeded in using the CO laser for obtaining 10-μm lasing from COF_2 and 8.6-μm lasing from $O^{13}CS$.

Doubled CO_2 Pump Laser

Frequency doubling the CO_2 laser allows all advantages of this laser except one: the peak power at the doubled frequency is limited by damage to the nonlinear crystal. However, the number of optical pumping wavelengths obtainable from the CO_2 laser is doubled by this technique, and these additional frequencies permit optically pumped emission in the 5 to 9- μm range, which is not available from normal CO_2 laser optical pumping.

Kildal and Deutsch[15,16] have obtained MIR laser emission from seven molecules through the use of doubled CO_2 laser optical pumping. All of these molecules lased as result of V-V collisional transfer from CO, which was excited by the second harmonic of a CO_2 laser line. The important type of laser exemplified here is termed the optically pumped transfer laser.

One of the primary merits of this kind of laser is that a very close spectral coincidence between the pump laser and the lasing molecule is not required. Instead, a match between the pump and the energy-storing donor molecule (e.g., CO) is necessary, but this coincidence can be as automatic as in the reported HBr-CO_2 (Ref. 12) and HF-CO_2 (Ref. 3) systems. Work on the second-harmonic CO_2 laser pumping demonstrated again that high-pressure (16 atm in the case of CO-CO_2) operation of type-B optically pumped laser is possible.

Also in the CO-CO_2 system a high conversion efficiency of 34% and an output energy of 13 mJ were measured. Note that, for reasons discussed previously, all of these seven molecules lased in the type-B mode.

CO_2 Pump Laser

By any criteria the CO_2 laser has been and is the most important pumping source for both MIR and FIR optically pumped lasers. Its capabilities of single-line operation on many wavelengths over the useful 9- to 11- μm range and high peak powers combine to offer the possibility of energetic and efficient optically pumped lasers. Its available output frequencies are in the spectral region of many molecular vibrational frequencies and allow generation of optically pumped emission from ~ 11 μm and beyond. For achieving lasing at wavelengths of less than 20 μm, cooling of the optically pumped medium is often helpful, since at room temperature the thermal population of the lower laser level can be significant.

Ten different molecules have lased in the MIR because of CO_2 laser pumping. Among the first molecules reported were CO_2 (Ref. 17), OCS (Refs. 18 and 31), and SF_6 (Ref. 19). In 1976 Chang and McGee[20] discovered the optically pumped NH_3 laser emitting at 12.812 μm, which today is the most powerful optically pumped laser. Later work has been reported on C_2H_4, NOCl, CF_3I, and stable isotopic forms of CF_4. Certainly, NH_3 has received the greatest amount of effort of any optically pumped MIR laser, but recently CF_4 has attracted considerable interest.

As one may expect, most of the CO_2 pump lines generating MIR lasing from NH_3 had previously been shown to produce FIR lasing. Among many interesting results on the NH_3 laser, Chang and McGee[22] presented strong evidence for the importance of two-photon, Raman-like processes for several lines effective in pumping NH_3 and C_2H_4. Fry[23] demonstrated the efficacy of rotational relaxation, subsequent to pumping a single level of NH_3, in producing many laser lines. Jacobs et. al.[24] showed that two pump photons can combine to pump ammonia and yield lasing in a wavelength region different from that achieved with single-frequency pumping. Certainly of major interest are the 1-MW output at 12.8 μm (Ref. 27) and large efficiencies reported for the CO_2-pumped NH_3 laser.[27,28]

The CF_4 laser discovered by Tiee and Wittig[26] is of interest for generating 16-μm radiation. They showed that several millijoules of energy could be generated somewhat easily, and they found a dozen lasing lines near 16 μm. Jones, Telle, and Buchwald[30] demonstrated lasing on all stable isotopes of this molecules - $^{12}CF_4$, $^{13}CF_4$ and $^{14}CF_4$ - and have produced over 80 lasing lines in the 16-μm region. For all three isotopic forms, they achieved numerous lasing lines on P, Q, and R branches on the $\nu_2 + \nu_4 - \nu_2$ transition and demonstrated the difficulty encountered in Q-branch lasing in a type-A system. They also have shown the importance of precision pump frequency control (not only single-mode operation) to the operation of the CF_4 laser, results which undoubtedly apply to most other optically pumped systems, particularly those not employing very high pump powers or large absorption transition moments.

IV. POTPOURRI

Many of the very important aspects of MIR optically pumped lasers will only be touched on here. For example, cavity design obviously has a large impact on the performance of these lasers. In addition to the normal design considerations of laser resonators, the method of introducing the pump beam into the signal cavity must be weighed. The optical configuration used must take into account details of the pumped medium; for example, type-A systems absorb pump radiation on a combination band. Because of the resulting small absorption coefficient, multiple pump passes or a very long cell must be used.

The pump frequency, vis-à-vis the lasing medium absorption spectrum, was discussed on a coarse scale in a previous section. Normally, a single line from the pump laser will be resonant with an absorption line of the lasing molecule to within 0.01 cm^{-1}. Often, details of the pump laser emission, of the absorbing line in the lasing molecule, and of the interaction between the two must be considered.

To cite an example, in the case of the TEA CO_2 pump laser, single-line emission consists of perhaps 20 individual frequencies - cavity modes - spanning up to $\sim$ 0.03 cm^{-1}. The longitudinal mode spacing may be on the order of 50 MHz, and in the absorbing molecule the linewidth may be $\sim$ 100 MHz. Thus, only a small fraction of the single-line pump power may be effective. Further, the pump laser modes will normally drift in frequency owing to thermal cavity length changes and will cause unstable lasing in the case of narrow absorption lines. Recent work demonstrates efficiency increases resulting from CO_2 laser mode selection[28] and from full frequency control.[30]

A fundamental problem facing many optically pumped lasers is intrapulse frequency chirp of the pump laser emission, a phenomenon that can affect the coupling between the excitation source and the absorbing medium. Details of the absorption features that can be important are exemplified in CF_4, for which diode spectroscopy has been employed to resolve spectra in the regions of pump and emission.[31] In this case, approximately five distinct absorption features are observed within each lasing linewidth of the single-line TEA CO_2 laser. Frequency control of this pump laser selects the proper absorption feature.

Other details are important at large pump intensities and/or large transition moments in the lasing molecule. Under these conditions the dynamic stark effect can broaden the absorption profile and have similar effects on the lasing transition.[32]

Also, stimulated Raman emission enhanced by near resonances have been shown to be important in some cases and offer prospects of greater efficiency than can be obtained in normal single-photon excitation. Further, one must consider saturation of the lasing medium absorption. Golger and Letokhov[33] have discussed conditions under which saturation of the pumped vib-rotation line and of the entire vibrational manifold occur.

V. CONCLUSION

Optically pumped lasers emitting in the mid-infrared region have exhibited a wide range of lasing wavelengths and a capability of high-power levels. In contrast with most coherent sources depending on nonlinear processes, there appear to be no general restrictions to the attainable output powers, either peak or average, from these lasers.

At this stage of development this laser allows line tuning over much of the middle infrared. However, only in the case of lasing from high-pressure CO_2/N_2O mixtures[13] has continuous tunability been demonstrated. Whereas at present this lasing system must be considered a special case, continuous tuning in other spectral regions will be available in the future. Laser pumping of appropriate condensed-phase species will be one path to this goal.

The state of the art in MIR optically pumped lasers is summarized in Fig. 4, where the wavelength ranges of standard mid-infrared lasers and of MIR optically pumped lasers are indicated. In the latter case each of the wavelength ranges can often be extended and gaps filled in by isotopic substitution in the lasing molecule. Using available spectroscopic data of gas-phase molecules, a species can be selected that will lase very near a prescribed frequency.

Finally, we note that mid-infrared lasers have already found applications. In 1976, Patel, Chang, and Nguyen[34] reported the use of the 12.8-μm ammonia laser to pump a spin-flip Raman laser to achieve a tunable output over the 14- to 16-μm range. Very recently, in separate work, the NH_3 laser has been used to dissociate

CCl_4 and SeF_6 molecules in an isotopically selective manner.[35,36] Advances in available frequencies and in optical power offered by mid-infrared optically pumped lasers will combine to substantially increase the use of these sources in the future.

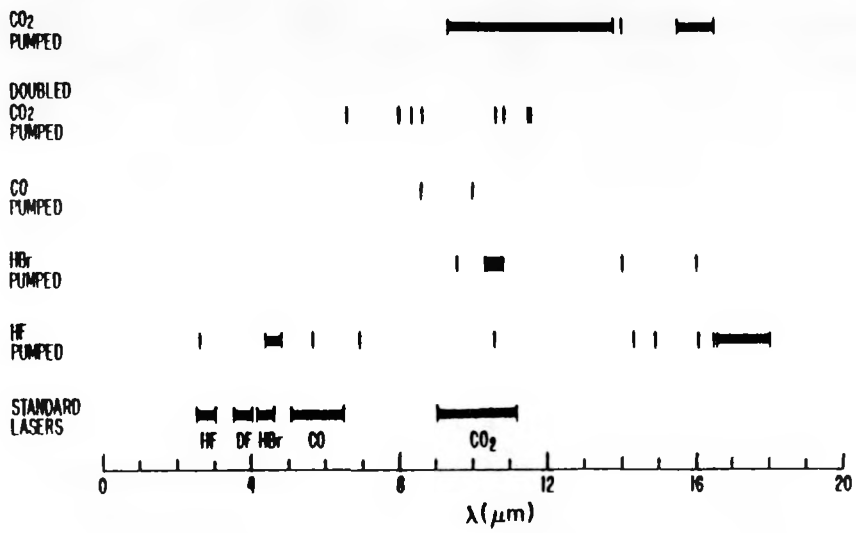

Fig. 4.
Wavelengths of lasing from standard mid-infrared lasers and from optically pumped lasers. Vertical bars indicate approximate wavelength for a particular laser, and the horizintal bars imply a somewhat dense spectrum over the indicated range. Only in the case of HBr-pumped CO_2/N_2O has continuous tuning been realized.

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