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OPTICALLY PUMPED FAR INFRARED LASERS

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INTRODUCTION

The search for efficient and powerful sources of coherent radiation spanning the widest possible spectral range is a major task in the fields of Quantum electronics and Laser spectroscopy. Until the discovery in 1970 [1] of the optically pumped Far Infrared (FIR) lasers, the submillimeter range of the spectrum had been almost barren due to the lack of radiation sources. Before 1970, the FIR portion of the electromagnetic spectrum was only very sparsely covered by molecular glow discharge lasers (HCN, DCN, H_2O) and the list of available laser lines was rather poor[2]
Since 1970 more than two thousand optically pumped FIR laser lines throughout the 0.03 to 2 mm region were discovered, some providing hundreds of Kilowatts of peak power in pulsed operation, others providing a few hundreds of milliwatts of continous power.
The emission lines are so closely spaced in the wavelength range between 1 mm and 0.03 mm that these sources of radiation can be considered to be step-tunable. Power fluctuation can be limited to a few percent and frequency stability can be better than 1 part in 10^9. In fact the emitted lines are in general homogeneously broadened and the linewidths of the resonator modes are larger than the gain curve of the active medium. As a consequence, the frequency of the emitted radiation is determined by the molecular transitions and is only slightly perturbed by the cavity tuning via pulling effects. This makes it possible to use FIR sources in high resolution spectroscopy without the need of active frequency

While for the pulsed regime other sources (FEL) are forecast, the optically pumped FIR laser will remain the only practical CW source for some years to come at least in the region 1 to 10 THz. A number of review papers were published in the past[3-9]. A critical review of the laser emissions from 30 of the most useful molecules has also been published[10] .

FIR LASER SYSTEM

The FIR lasers need as a pump source a laser emitting photons with an energy well above the KT in order to excite the molecules in an empty vibrational state. The laser action takes place between the rotational states of this vibrational manifold as shown in fig. 1.

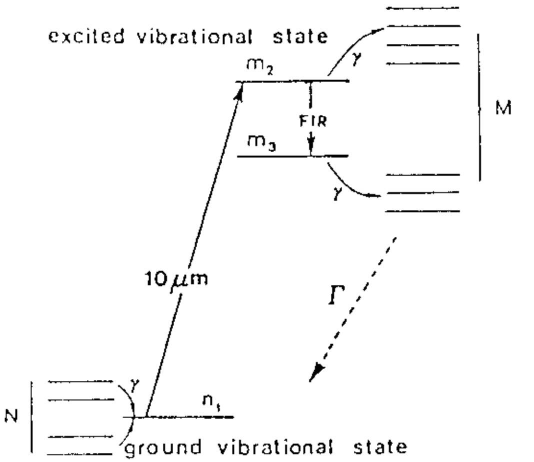

FIG. 1

The most used pump laser is the CO_2 , in some cases the isotopic CO_2 lasers, the CO_2 sequence lines laser and the N_2O laser were also used. A typical long pulse or continous-wave optically pumped FIR laser system is shown in fig. 2. The pump source must have an output power in the 3-60 W range. Only in the case of very efficient FIR laser lines the threshold pump power can be as low as a few hundred milliwatts. The pump laser is line selected by means of a diffraction grating while tuning resonance with the absorption line is obtained by a PZT tuner.

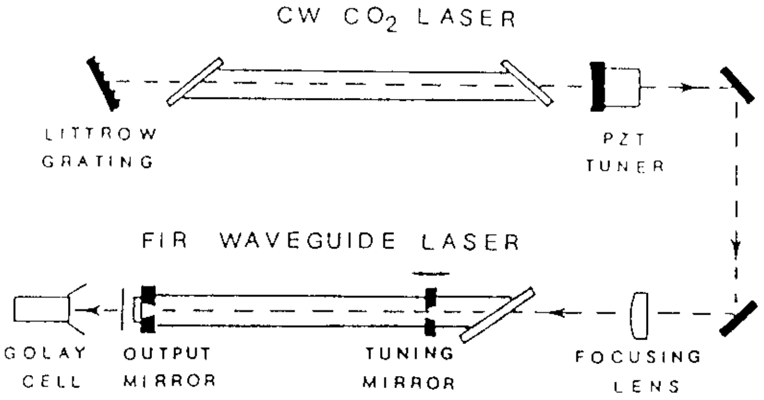

FIG. 2

In many cases a waveguide resonator is used for the FIR. A hollow dielectric cylindrical guide (a Pyrex tube) is a popular solution since the lowest attenuation mode is the EH_{11} , linearly polarized without a preferred direction[11] . The FIR electromagnetic field can be excited either parallel or orthogonal to the polarization of the pump radiation according to the ΔJ selection rule [3,4]. In the case of a cylindrical metallic waveguide the lowest attenuation mode is the TE_{01}. The electric field has a minimum on the axis and a circular polarization that misfits the pump radiation and causes a lower conversion efficiency. The rectangular hybrid waveguide consisting of two metallic and two dielectric sidewalls is also used since in this case a homogeneous electric field can be easily applied on the FIR laser active medium. By means of the Stark effect new FIR laser lines, a significant power enhancement and a moderate frequency tuning have been obtained for the most powerful CH_3OH laser lines [5].The waveguide resonators are terminated by flat mirrors that must be oriented orthogonal to the waveguide axis. Metal mirrors have a very good reflectivity at FIR wavelengths.
Also a confocal mirror cavity can be used. The fundamental mode is the TEM_{00} that combines very well with the pump radiation. The mirrors resonator is convenient for long wavelength FIR lasers ($\lambda > 0.5$ mm) since in this case it has a lower threshold pump

power than the W.G. lasers. On the other hand W.G. lasers have a better conversion efficiency and a larger output power. In fact the vibrational relaxation in the FIR laser is dominated by the molecular diffusion toward the wall wich depends critically upon the resonator diameter.

The pump laser power is coupled in the FIR laser by means of a small hole (d = 1 - 3 mm) at the center of the input mirror. A larger (d = 4 - 15 mm) hole in the output mirror is often used for extracting the FIR power. A variety of more sophisticated output devices have been used[7], however a hole of the proper diameter drilled in a metal mirror can always approach the best extraction efficiency independently of the FIR wavelength.

One mirror must be translatable along the laser axis in order to tune the FIR cavity in resonance with the FIR laser line. In fact the laser line is much narrower than the cavity free spectral range. The recording of the FIR output power as a function of the cavity length is a simple and convenient method to measure the FIR laser wavelength with a maximum precision of about one part in 10^4. Moreover in several molecules two, three or more FIR laser lines are obtained for each pump line. Again the separation of the different lines is obtained by changing the FIR cavity length.

RATE EQUATION MODEL

A rate equation model has proved to be quite useful in the description of the optically pumped FIR lasers[7,12,13]. The model is based on describing the kinetics as the sum of two separate laser interactions that share a common level and including all the collisional processes relevant for the relaxation of the perturbed states toward their equilibrium values. The rate equation model has proved to be in qualitative agreement with all the relevant FIR laser features with the obvious exception of effects like the AC Stark effect and the Raman gain that require a quantum mechanical treatment. A common lay-out in the description of the CW FIR laser takes into account that the pump transition is Doppler broadened while the FIR transition is homogeneously broadened (typical pressure broadening ~ 20-30 MHz/Torr). Moreover it is demonstrated experimentally[15,16] that the velocity and M changing collisions are negligible compared to the $\Delta J, \Delta K, \Delta V$ changing collisions. As a consequence of the Doppler broadening in the CW FIR lasers, only a fraction of the molecules in the level n_1 (fig. 1) can be simultaneously excited and the velocity dependent excitation rate is

$$W(\nu) = \frac{I_p(\nu)}{f_1 N}\,\alpha^{s}_{IR}(\nu)\,\exp\left[-4\ln 2\left(\frac{\nu-\nu_0}{\Delta\nu_D}\right)^2\right] \qquad 1)$$

where $\alpha^{S}_{IR}(\nu)$ is the saturated absorption coefficient for the pump transition, I_p the pump intensity, f_1 the fractional rotational equilibrium population and N the ground state vibrational manifold population. Since the IR absorption per transit in the FIR cavity is small we can assume that the pump radiation is propagating in the cavity with about the same intensity in both directions. In this case, as a first order expansion in I_p/I^S_{IR}

$$\alpha^{s}_{IR}(\nu) = \alpha^{0}_{IR}\left[1+\frac{I_p}{2I^{s}_{IR}}\left(1+\frac{(\Delta\nu_H/2)^2}{(\nu-\nu_0)^2+(\Delta\nu_H/2)^2}\right)\right]^{-1} \qquad 2)$$

where α^{0}_{IR}, I^S_{IR}, and $\Delta\nu_H$ are the unsaturated absorption, the saturation intensity and the homogeneous linewidth of the IR transition respectively. The relaxation between levels is a consequence of collisions. The rotational relaxation within a given vibrational state is very fast ($\gamma \sim 10^{10}$ $sec^{-1}atm^{-1}$) while the relaxation between the vibrational states is much slower ($\Gamma_c \sim 10^5$-10^7 $sec^{-1}atm^{-1}$). As a consequence also the wall collisions contribute significantly to the excited vibrational states relaxation, with a diffusion rate proportional to $p^{-1}d^{-2}$ (p: gas pressure, d: FIR resonator diameter). The spontaneous radiative emission being negligible we have

$$\Gamma = \Gamma_c \cdot p + \Gamma_D/p \simeq 10^3\text{-}10^4\ s^{-1}\,Torr + 10^3\ s^{-1}\,Torr^{-1}$$

In conclusion in the FIR laser there is a bottleneck with a fast thermalization within the vibrational states followed by a slower relaxation between the vibrational states. The fraction of molecules in the upper vibrational level is

$$M = M_p + M_e = \frac{f_1 N W(\nu)}{\Gamma} + M_e \qquad 3)$$

where M_e is the population at thermal equilibrium. At the steady state

$$\left(\frac{dm_2}{dt}\right)_p = f_1 N W(\nu) = -\left(\frac{dm_2}{dt}\right)_c = \gamma\left(m_2 - f_2 M\right)$$

and

$$m_2 - m_3 = \frac{f_1 N W}{\gamma} + M\left(f_2 - \frac{g_2}{g_3} f_3\right) \qquad 4)$$

The unsaturated gain for the FIR line is then

$$\alpha^{o}_{FIR} = h\nu_{FIR}\,\sigma_{FIR}\left[\frac{f_1 N W}{\gamma} + M\left(f_2 - \frac{g_2}{g_3} f_3\right)\right] \qquad 5)$$

where σ_{FIR} is the stimulated emission cross section and a homogeneous broadening has been assumed for the FIR transition. The FIR gain is positive only when

$$\frac{f_1 N W(\nu)}{\gamma} \geq M\left(\frac{g_2}{g_3} f_3 - f_2\right) \qquad 6)$$

which is a threshold condition for the FIR laser. When $M_p >> M_e$ eq. 6 becomes

$$1 \geq \frac{\gamma}{\Gamma}\left(\frac{g_2}{g_3} f_3 - f_2\right) \simeq \frac{\gamma}{\Gamma} f_2 \frac{h\nu_{FIR}}{KT} \qquad 7)$$

that in most cases cannot be satisfied without considering the diffusion relaxation. There is a cutoff pressure given by

$$p_c \leq \left[\frac{\Gamma_D}{\gamma'\left(\frac{g_2}{g_3} f_3 - f_2\right)}\right]^{1/2} \qquad 8)$$

where $\gamma = \gamma p$.

The typical operating pressure, around $p_c/2$, is in the range 0.3-0.03 Torr.

The output power from a homogeneously broadened laser is approximately given by

$$P_o \simeq V \frac{T}{\delta + T} \alpha^{o} I^{s} \qquad 9)$$

where V is the volume of the active medium, T the output mirror transmission and δ the round trip cavity losses. By using eq. 5

$$\frac{P_{FIR}}{V} = \frac{T I^{s}_{FIR}}{\delta + T} \cdot \sigma_{FIR} h\nu_{FIR} f_1 N W\left[\frac{1}{\gamma} - \frac{1}{\Gamma}\left(\frac{g_2}{g_3} f_3 - f_2\right)\right] \qquad 10)$$

where fNW(ν) can be conveniently written as

$$f_1 N W = \alpha^{s}_{IR} I_{IR} = \frac{P_{IR}}{V} \frac{\alpha_{IR} L}{\delta_{IR} + \alpha_{IR} L} \frac{1}{h\nu_{IR}} \qquad 11)$$

where L is the FIR cavity length and δ_{IR} the FIR cavity losses at IR frequency. Since

$$\sigma_{FIR} I^{s}_{FIR} = \gamma / \left(1 + \frac{g_2}{g_3}\right)$$

eq. 10) can be finally written as

$$\frac{P_{FIR}}{V} = \frac{P_{IR}}{V} \frac{\nu_{FIR}}{\nu_{IR}} \cdot \frac{\alpha_{IR} L}{\delta_{IR} + \alpha_{IR} L} \cdot \frac{T}{\delta_{FIR} + T}\left[1 - \frac{\gamma}{\Gamma}\left(\frac{g_2}{g_3} f_3 - f_2\right)\right]\left(1 + \frac{g_2}{g_3}\right)^{-1} \qquad 12)$$

The above equation represents a guideline for evaluating the maximum expected conversion efficiency of an optically pumped FIR laser. The meaning of the various terms in eq. 12 is straightforward. The frequency ratio expresses the quantum conversion efficiency multiplied by a factor of order 1/2 since $g_2 \simeq g_3$ and the rotational lifetime has been assumed equal for all the levels. P_{IR} is the pump power injected in the FIR cavity. The next two terms express the cavity efficiency at IR and FIR wavelengths respectively. The final bracketed term includes the molecular dynamics as discussed in eq. 5-8.

I^{s}_{FIR} and α^{o}_{FIR} do not appear in eq. 12 as a consequence of oversimplification of eq. 9, leading to the conclusion that all the lines of a given molecule would have the same efficiency. In fact it is possible to assume $2\alpha^{o}_{FIR} L >> \delta_{FIR} + T$ only in the case of high gain FIR lines. A less approximated eq. 9 must be used when the gain is low[11]. It is worth noting that even for the strong IR lines α^{o}_{IR} is of the order of unity in m^{-1} Torr^{-1}.

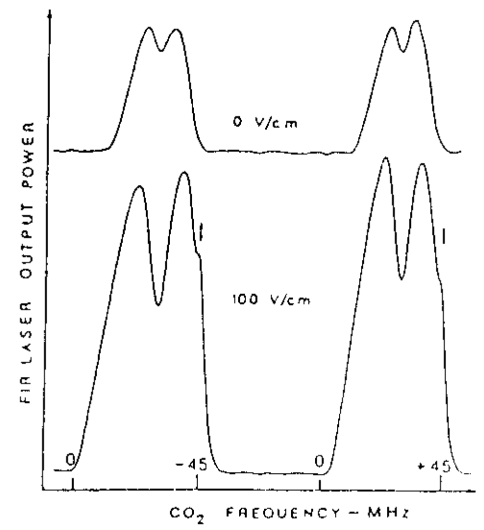

FIG. 3- Transferred Lamb dip observed in the 0.119 mm FIR laser line as a function of the CO_2 frequency tuning(after ref.24)

As a consequence it is important for an efficient conversion to have a FIR cavity with small IR absorption. The rate equation model can explain other observed effects. As an example from eq. 2 a saturation pump dip is expected when the pump laser frequency is tuned in resonance with IR absorbing line. As a consequence the FIR laser output must have a corresponding power decrease as is clearly shown in fig. 3. This effect is known as the transferred Lamb dip and is particularly useful for a precise measurement of the pump offset[5,17] .

SELECTION CRITERIA FOR THE FIR LASER LINES

Up to the present time FIR laser lines have been obtained from about 100 molecules. However the observed frequencies and the efficiency are quite different and some further criteria may be useful for the selection of the molecules.

Frequency

To a first order the rotational energy of a symmetric top molecule is

$$W_R = B J(J+1) + (A-B) K^2 \qquad 13)$$

The selection rules for rotational lines are $\Delta J = 1$, $\Delta K = 0$ and the emitted frequency is given by $\nu = 2BJ$. Since the B constant is inversely proportional to the moment of inertia the FIR laser lines are confined in the region 1 - 0.4 mm with the only exception of some special molecules like NH_3. In asymmetric as well as slightly asymmetric top molecules the permanent electric dipole moment may have a non vanishing component along all the principal axes of inertia and transitions with $\Delta K = \mp 1$ (or equivalent for K_{+1} and K_{-1}) are allowed adding a new energy term to the transitions. In the case of lightweight nearly symmetric top molecules like CH_3OH and CH_2F_2 the FIR laser lines are shifted mainly in the 0.4 0.1 mm region (fig. 4). A further effect like inversion or internal rotation may add a new energy term to the levels. The most important case is that of CH_3OH where the internal rotation of the OH group with respect to the CH_3 methyl group needs the introduction of two new quantum numbers τ, and n. As a consequence the number of levels populated at room temperature is increased by about a factor six and the selection rules are relaxed to $\Delta J = \pm 1, 0$, $\Delta K = \pm 1, 0$, Δn any. In particular the levels with n = 0 and n = 1 have an energy separation of about 150 cm^{-1} and the corresponding FIR laser lines a wavelength in the 0.1 - 0.03 mm region. For this reason the CH_3OH and its isotopically substituted forms are the most important molecules to obtain FIR laser lines with a wavelength in the 0.3 - 0.03 mm region.

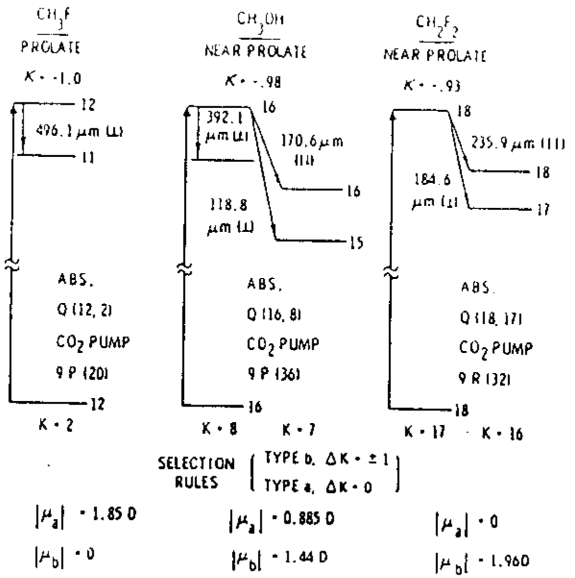

FIG. 4 - (After E.J. Danielewiecz in ref. 10)

Efficiency

Equation 12) is the starting point for selecting molecules candidate for high conversion efficiency FIR laser lines. In particular α^0_{IR} must be large, a condition satisfied when the electric dipole moment of the vibrational transition $|\mu_{IR}|$ is large and the rotational partition function Z is small. The ratio γ/Γ must be on the contrary as small as possible. The permanent electric dipole moment $|\mu|$ must be also large in order to have a large gain at the FIR transition. In table I are shown the values of the significant parameters for some of the most popular FIR lasing molecules. S is the band intensity in $cm^{-2}atm^{-1}$, Z the rotational partition function, X=S/Z, and Y=XZ (the data of table

TABLE I

MOLECULES	S	Z	X $(x10^3)$	Y $(x10^6)$
CH_3I	36	9294	4	3
CH_3F	440	2730	161	4.2
$C_2H_2F_2$	200	34772	6	30
CH_2F_2	1196	12404	96	159
CH_3OH	832	14280	58	1280
D_2O	400	225	2000	90000
NH_3	900	210	3000	170000

I are from ref. 7). The numbers in the last column are the guideline for the molecules selection. In fact the important molecule CH_2F_2 was successfully selected in this way[7,18] However it must be noted that a large number of coincidences with the CO_2 pump lines is possible only when Z is large. For this reason CH_2F_2 and CH_3OH are very rich in FIR laser lines while the more promising $^{14}NH_3$ have no coincidences with the CO_2 laser. Moreover not all the strong absorbing lines of a good molecule may produce strong FIR laser lines since the effective dipole moment $|\mu_{FIR}|$ is a function of the quantum numbers J and K and of the selection rules. As an example in the case of a symmetric top molecule the intensity of a IR line with $\Delta J=0$ (Q branch) is proportional to K^2 while the intensity of the associated FIR laser line is proportional to J^2-K^2. In table II are listed the most efficient FIR laser lines. The quoted powers have been obtained in different experimental conditions and are only indicative of the expected output power in optimized conditions and with an average pump power. The experimental evidence shows that the output power scales linearly with the pump power and the FIR cavity length. The conversion efficiency of the lines listed in Table II is of the order of 10~25%. This figure must be multiplied by the $\nu_{FIR}/2\nu_{IR}$ factor which reduces the absolute power proportionally to the FIR wavelength.

TABLE II

MOLECULE	CO_2 LINE	FIR (μm)	POL	POWER (mW)
$^{15}NH_3$	10R 18 ($^{13}CO_2$)	152.74	⊥	180
CH_3I	10P 18	477.15	//	40
HCOOH	9R 18	393.63	//	50
CH_2F_2	9R 32	184.30	⊥	150
	9R 20	117.73	⊥	70
CH_3OH	9P 36	118.83	⊥	400 (750^a)
	9R 10	96.52	//	300
	9P 34	70.51		100
	9P 32	42.16	//	50
	9P 16	570.57	//	38
CD_3OH	10R 18	41.35	⊥	60
$^{13}CD_3OH$	10P 8	127.02	⊥	b

a- High pumping power; see ref. 20

b- Stronger than the CH_3OH 119 μm; see ref. 29

IMPROVING THE EFFICIENCY OF THE LASERS

The output power of the optically pumped FIR laser can be improved in several ways.

1- Cavity length, pump power

By increasing the FIR cavity length the quantity of lasing molecules incresases proportionally, thus increasing also the output power as long as the single pass attenuation of pump power remains small. Experimentally a power increase proportional to the cavity length has been observed up to 3-4 m in the case of CH_3OH 119 and 71 μm laser lines. In this way it is possible to bypass the vibrational relaxation bottleneck discussed in the previous section. The limit in the useful cavity length depends also on the available pump intensity and on the pump saturation effect.

2- CO_2 tunability

The gas absorption depends on the frequency detuning of the CO_2 laser from the absorbing line center. FIR line emission can be usually observed only for detunings smaller than the Doppler linewidth as shown in fig. 3. Since a perfect coincidence with the CO_2 line is unlikely, a CO_2 laser with a TEM_{00} mode frequency tunable over all the free spectral range is more convenient than a higher power but multimode laser that cannot be tuned as a consequence of mode hopping. In many cases FIR laser lines classified as weak were found to be much stronger when pumped by a short cavity CO_2 laser with less power but larger tunability.

3- FIR temperature

From the rate equation model the conversion efficiency is proportional to the number f_1N of molecules in the ground state. The fraction f_1 is a function of the gas temperature and can be maximized either by heating or cooling the FIR laser. As an example both the 496 μm line of CH_3F and the 119 μm line of CH_3OH have an optimum temperature around 0°C with a power increase of about 30% with respect to the room temperature 19,20,8.

4- Buffer gases

The VV and VT relaxation rates can be increased by adding a buffer gas. A lightweight and transparent buffer gas like H_2, and He is useful in cooling the gas that is heated by the absorbed pump power and by increasing the V-T relaxation rate. In fact a power increase of about 30-50% was found in the case of the 119 μm CH_3OH laser [20]. On the contrary no beneficial effect was observed in the case of the 496 μm CH_3F laser[21,9]. A power increase of about 80 % was observed for this laser when SF_6 was used as a buffer gas[22](fig.5). In this case there is a very fast near resonant VV transfer between the two molecules and the SF_6 has also a fast VT deactivation [9]. The SF_6 gas has proved to produce the greatest increase in output among all the buffer gases reported. Unfortunately it is also a strong absorber and must be used only with a fraction of CO_2 laser lines.

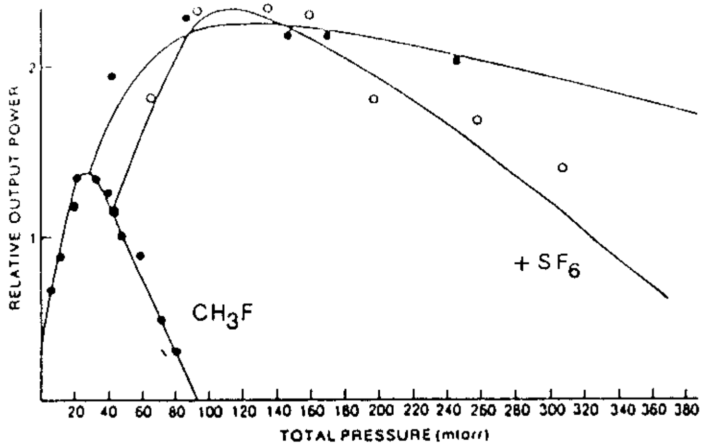

FIG. 5 - Relative power emitted by 0.496 mm CH_3F FIR laser as a function of the total gas pressure with SF_6 as a buffer gas (after ref. 22)

5- Two photon pumping

In some cases the ground state is connected to the other states of the ground vibrational manifold by microwave or radiofrequency transitions. The pumping rate can then be increased by transferring molecules from these levels by means of a resonant MW or RF pump field (fig. 6). An output power increase was also observed when the RF field was used to depopulate the lower FIR laser level (fig. 6). The two effects are cumulative and can be used to obtain a power increase of the order of 15-30% [23].

6- Non linear Hanle effect (NLHE)

The molecules candidates for FIR laser sources must be polar molecules with a large permanent electric dipole moment. For many of them, the Stark effect splitting of the M sublevels is larger than the homogeneous linewidth even when the applied electric

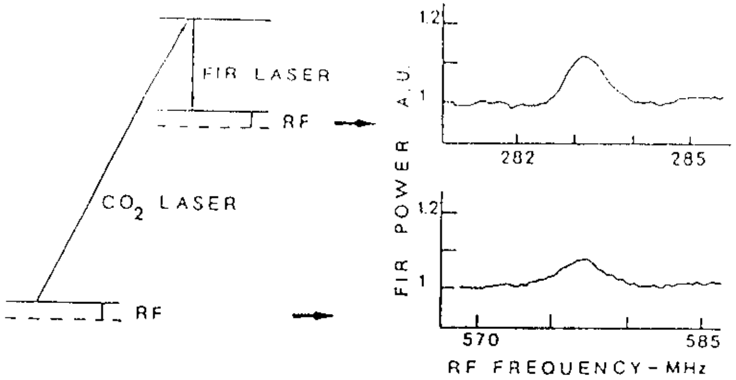

FIG. 6 - Output power increase of the CH_3OH 251 μm FIR laser with resonant RF pumping (after ref.23)

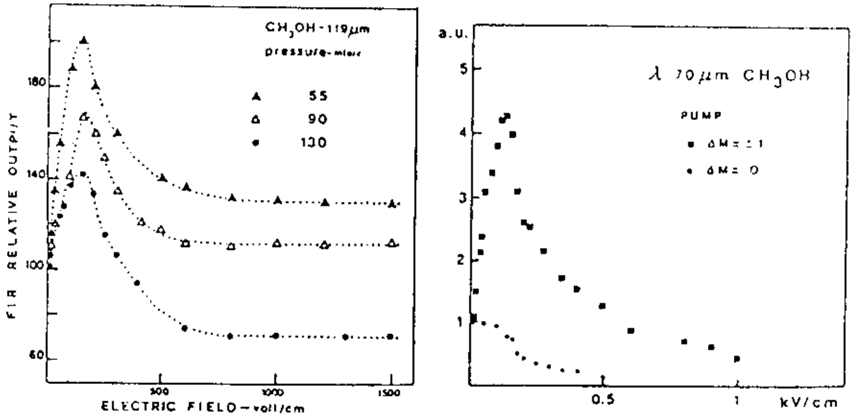

FIG. 7 - NLHE power enhancement effect as a function of the applied electric field (after ref. 24)

field intensity is of the order of a few hundred V/cm. In this case a large power enhancement can be obtained for many FIR laser lines when the polarization of the CO_2 pump radiation is orthogonal to the Stark field. Such power increase is explained in terms of the non linear Hanle effect (NLHE)[6,24,25] that predicts a saturated absorption increase for any transition when the degeneracy between ΔM=+1 and ΔM=-1 transitions is removed with respect to the homogeneous linewidth by an external field. In the case of the FIR laser the increase in the pumping rate may be of the order of 30-40 % and the increase in the FIR output power is as large as 100 % to 500% as shown in fig. 7. The NLHE, being a coherent effect between the M sublevels, is independent of the frequency tuning of the CO_2 laser around the IR absorbing line. As a consequence the power enhancement can be observed independently of the detuning and the lasing frequency interval is increased as is clearly shown in fig. 3. The NLHE enhancement has been observed in the case of many laser lines including all the methyl alcohol lines reported in table II.

It is worth to note that the effects described act independently of the FIR laser and as a consequence can be used simultaneously in order to obtain a substantial power increase. As an example methods 3 and 4 nearly doubled the output power of a 2.7 meters long 119 μm CH_3OH FIR laser[20] .

FREQUENCY TUNING

The FIR lasers are nearly fixed frequency laser sources. However the frequency tuning of some lines has been increased by about two orders of magnitude by using the linear Stark effect [6]. A fast frequency modulation (1 MHz) was also obtained[6] . For many powerful methyl alcohol FIR laser lines the Stark effect appears as a splitting in only two laser lines at respectively lower and

TABLE III [a)]

Line (μm)	CO_2 pump	Pump offset (MHz)	Tuning (MHz/kV cm^{-1})	Stark lines
37.9	9-*P*(32)	−16	59	2
70.5	9-*P*(34)	+36	44.2	2
96.5	9-*R*(10)	0	12	2
99.3	9-*P*(36)	−89	29	2
110.8	9-*P*(36)	−80	24.6	2
118.8	9-*P*(36)	+29	26.4	2
133.1	9-*P*(24)	+6	101	2
193.1	9-*P*(38)	+13	34	2
205.6	9-*P*(34)	+120	35	4
208.3	9-*P*(34)	>130	135	2
261.5	9-*P*(12)	+85	18.3	2
292.2	9-*P*(38)	+13	20.5	2

a) After ref. 6

higher frequency than that of the unperturbed line, as a consequence of the molecular selection rules (transitions with $\Delta J = \Delta K = 1$ and $K \simeq J/2$). In table III are reported the observed Stark tunings.

NEW EFFICIENT FIR LASER LINES

New FIR lasing molecules are continuously reported, however efficient molecules satisfying the selection criteria previously discussed are very few. New efficient laser lines are more likely to be found by using isotopic substitutes of already well known molecules like CH_3OH, NH_3, CH_2F_2. A similar approach is to use the isotopic CO_2 laser as a pump source. Recently powerful waveguide CO_2 lasers have been developed with an extended tunability with respect to the conventional one. In this way new coincidences with strong absorbing lines of "efficient molecules" have become available and a new set of strong FIR laser lines has been discovered in CH_3OH, $^{13}CH_3OH$, CH_2F_2 and many others will be available in the future[26,27,28].

REFERENCES

1. T.Y. Chang, T.J. Bridges, Opt. Commun. 1, 423 (1970)
2. P.D. Coleman, IEEE J. Quant.Electron. QE 9, 130 (1973)
   G.W. Chantry, G. Duxbury, in " Methods Exp. Phys.", vol. 3A, L. Marton ed., Academic Press, 1974 pagg. 302-394.
3. T.Y. Chang, " Optical Pumping in gases" in Y.R. Shen " Nonlinear Infrared Genration" Springer Verlag, Berlin, Heidelberg, New York, 1977, pagg. 215-272.
4. T.A. De Temple,"Pulsed optically pumped far infrared lasers", in "Infrared and millimeter waves" vol.1, K.J. Button ed., Academic Press, New York, pagg.129-184 (1979)
5. J.O. Henningsen,"Molecular Spectroscopy by far infrared laser emission",ibid., vol. 5, pagg. 29-129 (1982)
6. F. Strumia, M. Inguscio," Stark Spectroscopy and frequency tuning in optically pumped far-infrared lasers" ibid.,vol.5, pagg. 130-214 (1982)
7. T.A. De Temple, E.J. Danielewicz, " Continuous wave optically pumped lasers", ibid., vol 7, pagg. 1-41 (1983)
8. K. Walzer, Optimization of optically pumped far-infrared lasers,ibid. vol.7, pagg. 119-163 (1983)
9. J.P. Picharuthu, Submillimeter lasers with electrical, chemical and incoherent optical excitation.ibid. vol.7, pagg. 165-244 (1983)
10. K.J. Button, M. Inguscio, F. Strumia eds., Optically pumped far-infrared lasers, "Reviews of infrared and millimeter waves" , vol.2, Plenum Press, New York (1984)
11. F. Strumia,N. Ioli, High power, tunable waveguide CO_2 lasers, this book.
12. J.R. Tucker, IEEE Trans. Microw. Theory Tech., MTT-22, 1117 (1974)
13. D.T. Hodges, J.R. Tucker, Appl.Phys.Lett. 27, 667 (1975)
14. D.T. Hodges, Infr. Phys. 18, 375 (1978)
15. T. Oka in "Advances in Atomic and Molecular Physics", D. Bates, B. Bederson eds., vol. 9; Academic Press, New York (1973)
16. M. Inguscio, F. Strumia, K.M. Evenson, D.A. Jennings, D.A. Scalabrin; S.R. Stein, Opt. Lett. 4, 9 (1979)
17. M. Inguscio, A. Moretti, F. Strumia, Opt. Commun. 30, 355 (1979)
18. E.J. Danielewicz, C.O. Weiss, IEEE J. Quant. Electron: QE14, 705 (1978)
19. K.Walzer, M. Tacke, IEEE J. Quant. Electron. QE16, 255 (1980)
20. D.K. Mansfield, L.C. Johnson, R. Chouinard, Conf. Digest 8th Int. Conf. on Infr. Millim. Waves, Miami 1983, paper W5.4 IEEE cat. N° 83 CH 1917 - 4.
21. T.Y. Chang, C. Lin, J· Opt. Soc. Am. 66, 362 (1976)
22. N.M. Lavandy, G.A. Koepf, Opt. Lett. 5, 336 (1980)
23. N. Ioli, A. Moretti, G. Moruzzi, P. Roselli, F. Strumia, J. Mol. Spectr. 105, 284 (1984)
24. M. Inguscio, A. Moretti, F. Strumia, IEEE J. Quant. Electr. QE 16, 955 (1980)
25. F. Strumia, J. de Physique-Colloque C7, 117 (1983)
26. M. Inguscio, N. Ioli, A. Moretti, G. Moruzzi, F. Strumia, Opt. Commun. 37, 211 (1981)
    M. Inguscio, F. Strumia, J.O. Henningsen, in ref. 10
27. M. Inguscio, N. Ioli, A. Moretti, F. Strumia, "New FIR laser emissions in CH_3OH" Proc. Third CIRP Conf. Zurich 1984,p.448
    N. Ioli, A. Moretti, G. Moruzzi, F. Strumia, F. D'Amato, "New large offset FIR laser lines in $^{13}CH_3OH$, $^{13}CD_3OH$ and CH_2F_2" Proc. Ninth Int. Conf. Infrared and Millim. Waves Osaka 1984
28. G. Merkle, J. Heppner, Opt. Commun. in press (CH_3OH) and Opt. Lett. in press (CH_2F_2)
29. M. Inguscio, K.M. Evenson, F.R. Petersen, F. Strumia, E. Vasconcellos, Int . J. Infr. and Millim. Waves, in press.