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High Efficiency cw Far Infrared Lasers at 119 µm and 127 µm

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Abstract. The optically pumped FIR laser lines at 119 µm from CH_3OH and at 127 µm from $^{13}CD_3OH$ are known to be the most powerful in the far infrared spectral region. We report on efficiency measurements for our waveguide laser system. The effect of various parameters was investigated, resulting in the highest efficiency ever reported for the 119 µm line. The Stark effect and others parameters of the 127 µm were measured, and a new $^{13}CD_3OH$ laser line at 175 µm discovered, with the same pump transition. These measurements are helpful for completing the assignment already proposed for the 127 µm line.

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The development of efficient, high output power FIR lasers is of particular interest for many practical applications. Powerful laser lines in the region 200–100 µm are crucial for plasma diagnostics, especially for the large new tokamak machines. The optically pumped CH_3OH molecule has proven to be the most successful molecule for the generation of efficient and short wavelength laser lines. In particular the 119 µm laser line, pumped by the $9P(36)$ CO_2 laser line, is among those exhibiting the highest quantum efficiency in the FIR, and has delivered the highest power reported [1–3]. However the maximum power for this line represents only 16% of the maximum attainable power expected from the Manley-Rowe condition. This rather low quantum conversion factor leeves open the chance of discovering new, more efficient laser lines, possibly by following the criteria suggested by the rate equation model for optically pumped FIR lasers [1]. In fact a new, very promising line at 127 µm was discovered by investigating the isotopically substituted molecule $^{13}CD_3OH$ [4]. This line was pumped by the $10P(8)$ CO_2 laser line, more powerful than the $9P(36)$, and showed a larger efficiency than the 119 µm line in a direct comparison by using an open structure FIR resonator with a lateral output coupling [4]. Since it is well established that a dielectric waveguide resonator is more efficient for FIR wavelengths shorter than 200 µm, we decided to repeat the comparison by using a FIR laser designed for the maximum efficiency.

1. Experimental Apparatus

The FIR waveguide laser was made of Pyrex, 144 cm in length, and 4.0 cm in internal diameter. We have also used, for comparison, a copper waveguide 144 cm in length and 4.0 cm internal diameter. Both waveguides were alternately mounted in the same Super Invar mechanical frame to ensure the maximum mechanical and thermal stability. The resonator was terminated with flat copper mirrors, one with a 2 mm hole on the axis for the input of the pump CO_2 laser beam, the other with an off axis hole 12.8 mm in diameter for the output of the FIR radiation. The input hole was sealed with an AR coated ZnSe window and the output hole with an uncoated *z*-cut quartz window 1 mm thick, which is expected to transmit only 70% of the FIR power at 120 µm. All the power values reported in this paper are the values as measured outside the output window in the case of the FIR radiation and before the input window in the case of the CO_2 pump radiation. The powers of the FIR and CO_2 beams were measured with a Scientech 362 power meter that, in the FIR, was calibrated with a self-calibrating cone power meter as described in [5]. The absolute accuracy in the power measurement is expected to be of the order of a few %, and our calibration factor ($77 \pm 5\%$ efficiency at 119 µm) is in agreement with that reported in [6] ($80 \pm 10\%$ efficiency at 119 µm and about 75% at 127 µm). The commercial power meter was used routinely because of its faster response time.

The waveguide FIR laser was also modified for the investigation of the Stark effect. In this case a hybrid waveguide rectangular in cross section was inserted in the pyrex tube, as described in [7, 8]. The laser length remained unchanged and the dc voltage across the active medium was provided by a calibrated power supply connected to the metal walls of the hybrid waveguide. As a pump we used a waveguide cw CO_2 laser developed in our laboratory [9, 10].

2. Experimental Results

The offset between the CO_2 pump laser frequency and the center of the line pumping the 127 μm laser line, previously unknown, was preliminarily measured by using the accurate transferred Lamb-dip technique [11] as shown in Fig. 1. The CO_2 pump laser is frequency tuned across the cavity free spectral range and the corresponding output power of the FIR laser is recorded. Because the IR absorbing transition is Doppler broadened, and the FIR line is homogeneously broadened, the power decrease of the FIR laser at the exact pump coincidence represents the saturation Lamb dip in the pump transition [11], while no saturation Lamb dip is possible in the FIR laser line. We observed a nearly perfect coincidence between the CO_2 pump laser frequency and the $^{13}CD_3OH$ absorption line, as reported in Table 1, together with the already known pump offset (+24 MHz) of the 119 μm CH_3OH line. This result stresses the convenience for the 127 μm line, especially when a powerful CO_2 pump laser with a long cavity and a limited frequency tuning is to be used for obtaining the maximum FIR laser power.

The comparison of the output power obtained for the two FIR laser lines as a function of the CO_2 pump power is shown in Fig. 2. The pump power threshold was about the same for the two lines (≈3 W), but the 119 μm line was more powerful (slope: 58.5 mW for 10 W of pump power) than the 127 μm (23 mW for 10 W). The best operating pressure was about 12 Pa and 17 Pa respectively for CH_3OH and $^{13}CD_3OH$.

The observed quantum efficiency was estimated to be 14.3% and 5.45% respectively, and increased to 20.4% and 7.8% respectively if corrected for the output window transmission. The efficiency we obtained for the 119 μm laser line is the highest reported up to now, as shown in Table 2, column 6. In the table we have compared the best published results by reporting the FIR power obtained per unit of pumping power and the FIR power obtained per unit of pumping power and per unit of the FIR laser cavity length. In this way the experimental results obtained with different experi-

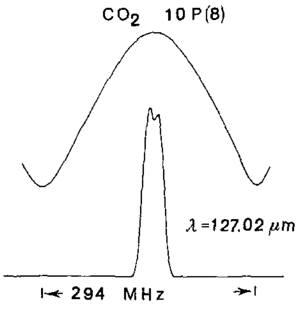

Fig. 1. Transferred Lamb-dip signal of the 127 μm laser line. The upper curve shows the CO_2 laser power as a function of its frequency tuning around the 10P(8) line and the lower curve the corresponding output power of the 127 μm line

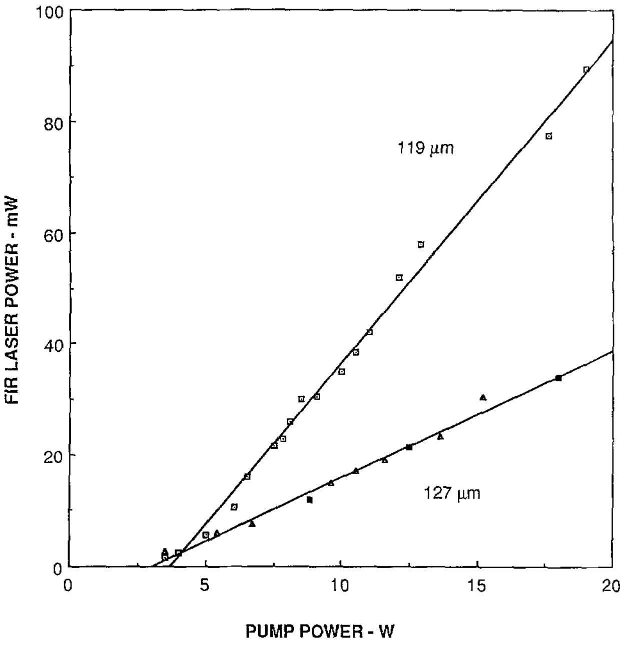

Fig. 2. FIR laser output power as a function of the CO_2 pump power. The measurements were performed by using the dielectric waveguide with the exception of that marked with ■, which was obtained with a copper waveguide of the same dimension

Table 1. Comparison of the experimental data of the 119 μm (CH_3OH) and 127 μm ($^{13}CD_3OH$) far infrared laser lines. The quantum efficiency is not corrected for the output window transmission. Data for the 175 μm new laser line from $^{13}CD_3OH$ are also reported

Wave [μm]	Frequency [MHz]	Pump line	Rel. Pol.	Offset [MHz]	Quantum Eff.%	Pressure [Pa]	Stark splitting [MHz/kV cm^{-1}]
118.8	2522781.6(5)	9P(36)	⊥	+24(2)	14.3	12	26.2
127.0	2348438.4(10)	10P(8)	∥	0(3)	5.45	17	22.8
175.26±0.1		10P(8)	⊥	0(3)		4	new line

Table 2. Comparison between efficient 119 μm dielectric waveguide lasers. Resonator parameters: L and ID are the length and internal diameter of the waveguide respectively; A.C. is the diameter of the output coupling hole. In the last three columns are given the output power for 10 W of pump power, the output power for 10 W of pump power and per unit cavity length, the output power for 10 W of pump power at the maximum pump power with He as a buffer gas

Ref.	L [m]	ID [cm]	A.C. [cm]	mW/10 W	mW/(10 W × m)	Max
This work	1.44	4.0	1.2	58.5	40.6	
[3]	2.5	3.4	1.0	76	30.4	40
[2]	2.67	3.0	1.0	73	27.3	24
[12]	2.0	3.4	1.0	54	27	

mental apparatus can be reasonably confronted. In fact the rate equation model of the FIR lasers predicts the power conversion efficiency as

$$\frac{P_{\mathrm{FIR}}}{V}=\frac{P_{\mathrm{IR}}}{V}\frac{\nu_{\mathrm{FIR}}}{\nu_{\mathrm{IR}}}\left(1+\frac{g_2}{g_1}\right)^{-1}\frac{\alpha_{\mathrm{IR}}L}{\delta_{\mathrm{IR}}+\alpha_{\mathrm{IR}}L}\frac{T}{\delta_{\mathrm{FIR}}+T}\times\left[1-\frac{\gamma}{\Gamma}\left(\frac{g_2}{g_1}f_3-f_2\right)\right], \tag{1}$$

where V is the active volume of the FIR laser, g_2 and g_1 the Landé factors of the levels, L the length of the FIR cavity, α_{IR} the absorption coefficient of the IR pump transition, δ_{IR} and δ_{FIR} the FIR cavity losses at IR and FIR wavelengths respectively, T the transmission of the output mirror, γ and Γ the rotational and vibrational relaxation rate respectively, and f_3 and f_2 the fractional rotational level populations at thermal equilibrium [1, 13].

The meaning of the various terms is straightforward: the first three terms represent quantum conversion limit, the next two terms express the cavity efficiency at IR and FIR respectively, the final bracketed term includes the molecular dynamics dominated by the vibrational bottleneck. The saturation and the small signal gain do not appear in the equation, as a consequence of the simplification

$$2\alpha^0_{\mathrm{FIR}}L \gg \delta_{\mathrm{FIR}}+T \tag{2}$$

which is valid only in the case of high-efficiency FIR laser lines (α^0_{FIR} is the unsaturated gain of the FIR laser). It is worth noting that, in contrast, α_{IR} is of the order of unit in $\mathrm{m}^{-1}\,\mathrm{Torr}^{-1}$, even for the strongest IR lines. Since the operating pressure is limited to a few hundreds mTorr as a consequence of the vibrational bottleneck, a large fraction of the pump power will be absorbed by the FIR cavity. As a consequence a FIR cavity with a small IR absorption is very important for an efficient power conversion. In practice $\delta_{\mathrm{IR}} > \alpha_{\mathrm{IR}}L$ and the output power is proportional to the length of the FIR cavity as it has been demonstrated experimentally.

At very high pump intensities non-linear effects become important: the temperature increases in the active gas and, as a consequence, increases also the population of other excited vibrational states. As a general feature the proportionality between the FIR power and the pump power becomes less than linear, but the output power can be increased by the temperature control of the FIR cavity and by the addition of He as a buffer gas. These improvements are ineffective with a low or moderate pump power and were not adopted in our experiment. For the same reason in Table 2 the efficiencies as observed at medium pump power are compared in column 6, while those observed at the largest pump powers and in presence of a buffer gas are reported in column 7. As a further comment it is worth noting that in our experiment the output window was made of uncoated crystal quartz, with a reflectivity of only 33% for the 9P36 $\mathrm{CO_2}$ radiation [14]. On the other hand, in [3] and [12] the crystal quartz output window was coated for a maximum reflectivity of the $\mathrm{CO_2}$ radiation. In [2] a high reflection coated Si window was used.

The lower efficiency observed in the case of the 127 μm line is in contrast with the previously reported data [4], obtained with an open structure near confocal resonator and a metal mirror coupler. A partial explanation of the present result can be related to the smaller reflectivity of the uncoated crystal quartz at the 10P8 wavelength: about 14% [14]. Therefore the IR cavity losses are increased according to the (1). We can estimate that this effect may reduce the power of the 127 μm laser line by about 25–30%. Another possible explanation for smaller 127 μm efficiency observed in our waveguide laser could be an unoptimized output coupling. In fact, our cavity was optimized for the 119 μm line but could be under or over coupled for the 127 μm line.

We have also used the copper waveguide obtaining the same power as for the Pyrex waveguide in the case of the 127 μm line (Fig. 2). This result is possible because this laser line is polarized parallel to the $\mathrm{CO_2}$ pump radiation so that the same transverse mode can be excited. In this case, however, the FIR laser beam is circularly polarized, a feature not convenient in most cases.

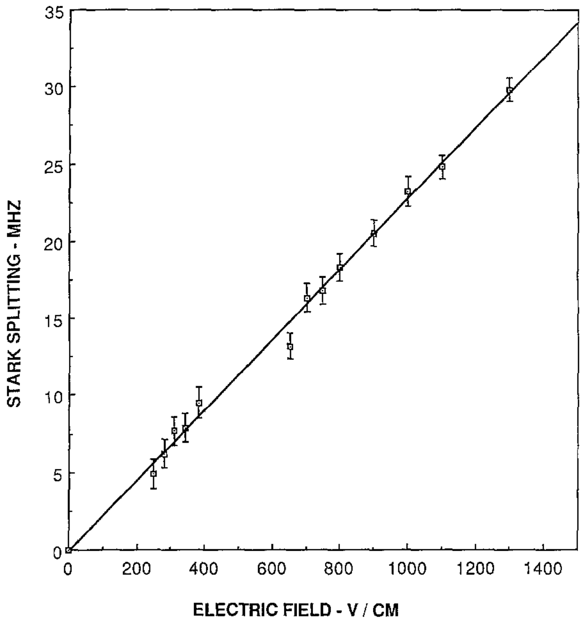

Fig. 3. Observed Stark splitting into two components for the 127 μm laser line

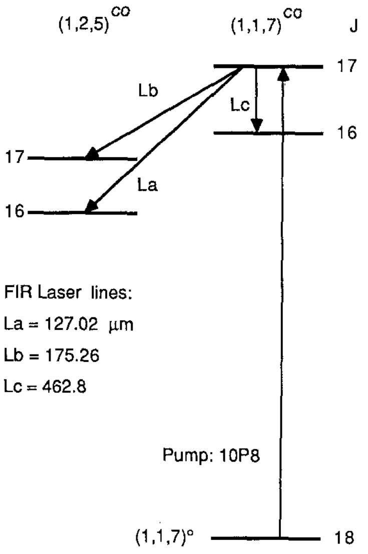

Fig. 4. Assignment scheme for the FIR laser lines from $^{13}CD_3OH$ obtained using the 10P(8) pump line

A frequency tuning and a fast frequency modulation of a FIR laser are important for many applications, such as for interferometric systems designed for the plasma diagnostic studies. Taking in account the results previously obtained for the 119 μm line, we decided to investigate the effect of a dc electric field on the 127 μm laser line active medium. Also for this line we discovered that the Stark effect splits the line into two components symmetric with respect to the unperturbed frequency. This feature is typical of the laser lines corresponding to a strong molecular transitions with the selection rules ($J \rightarrow J-1$; $K \rightarrow K-1$). In fact the frequency shift of each Stark component is given by the equation

$$\Delta\nu = \frac{\mu E}{J(J-1)}\left(\frac{2K-J-1}{J+1}M-(K-1)\Delta M\right), \tag{3}$$

where the first term in the bracket becomes negligible when $2K \approx J$ and all the M components collapse into only two close packets corresponding to the $\Delta M = \pm 1$ selection rule [7, 15]. The measured Stark splitting is shown in Fig. 3 and reported in Table 1. Recently, a possible assignment for the 127 μm laser line has been proposed [17] as an A-symmetry transition with the quantum numbers $(n, K, J) = (1, 6, 17) \rightarrow (1, 5, 16)$ and pumped by the P(18) IR line. This assignment is in qualitative agreement with the observed Stark effect; moreover, from [Ref. 7, Fig. 20], the Stark components with the largest laser gain correspond to $M = +17$, $\Delta M = +1$ and $M = -17$, $\Delta M = -1$. The parallel component of the electric dipole moment of $^{13}CD_3OH$ is not known but it can be assumed to be about the same as that of CH_3OH, namely $\mu \approx 0.9$ Debye. The Stark splitting expected from (3) is then $2\Delta\nu = 35.5$ MHz/(kV cm^{-1}), apparently not in good agreement with the experimental result. A more refined theory must take into account a possible difference in the electric dipole moment of the upper and lower laser levels, especially in the case of torsionally excited states. In this case the Stark frequency shift is given by the equation

$$\Delta\nu = \frac{-E}{J(J+1)} \times \left\{\mu M + \frac{KM}{J+1}\left[(J-1)\Delta\mu - 2\mu\right] + \mu(K-1)\Delta M\right\}, \tag{4}$$

where $\Delta\mu = \mu' - \mu$, μ' being the dipole moment of the upper laser level [16]. In contrast, the contribution to the Stark effect of terms quadratic in the electric field is negligible. The experimental value of the Stark splitting can be obtained from (4) by assuming

$$\Delta\mu = -0.029, \tag{5}$$

a value reasonable for methanol. In fact also in the case of the 96 μm laser line of CH_3OH pumped by the CO_2 9R10 line the observed Stark splitting is in agreement with the theory only assuming $\Delta\mu = -0.03$ Debye [16]. To confirm the proposed assignment, we also searched for the third laser line of the expected triad (Fig. 4), and we discovered a new line polarized orthogonally with respect to the polarization of the CO_2 pump radiation, as expected, with an output power about an order of magnitude smaller than that of the 127 μm line (Table 1). The measured wavelength of 175.26±0.1 μm is in good agreement with the expected value of 175.1 μm. In conclusion, there is a strong experimental evidence in favor of the correctness of the proposed assignment.

3. Conclusions

We have investigated the efficiency of the 127 μm and 119 μm laser lines. In the case of the second one we have obtained the largest known specific efficiency. The 127 μm laser line was found to be less efficient than the 119 μm line, in contrast with a previous experiment [4]. A possible explanation for this contradictory result was discussed. Other parameters of interest for applications, such as the pump offset and the Stark effect have been measured, and the results demonstrate that the 127 μm laser line can be an advantageous alternative to the 119 μm line. A new laser line was discovered, whose wavelength and relative polarization confirm the assignment of the 127 μm line as proposed in [7]. From the assignment, we predict that the efficiency of this line should further increase by increasing the gas temperature.

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