LRP 443/91 November 1991

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accepted for publication in

International Journal of
Infrared and Millimeter Waves

NEW CASCADE LASER TRANSITIONS IN CH_2F_2 PUMPED WITH THE 9R32 LINE OF A CW CO_2 LASER

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Abstract

New cascade laser transitions of $^{12}CH_2F_2$ at 172.50μm, 208.83μm, 220.44μm, 223.99μm and 250.61μm are reported. A waveguide FIR laser was pumped with a quasi cw $^{12}C^{16}O_2$ laser operating on the 9R32 line. Together with the already known lines at 184.3μm, 196.1μm and 235.9μm, the laser lines can be assigned to rotational transitions in the v_9 vibrational band of $^{12}CH_2F_2$ and to refill transitions of the vibrational ground state v_0.

Introduction

The 184.3μm laser line of CH_2F_2 was shown to be one of the most powerful FIR laser lines obtained by optically pumping with a CO_2 laser [1]. A cascade transition at 196.1μm was discovered and it was shown that it can contain 25% of the total output power of the FIR laser [2,3]. Furthermore a transition at 235.9μm was observed having its origin in the same upper level as the prominent 184.3μm line. In this paper some new laser lines at 172.50μm, 208.83μm, 220.44μm, 223.99μm and 250.61μm are reported. An assignment to rotational transitions of the $^{12}CH_2F_2$ molecule in the ν_9 band was made following Danielewics [4] and Davis [2].

Experiment

The CO_2 laser used for pumping was an Edinburgh Instruments PL4 model with a maximum power of 56W on the 9R32 line. It was operated in a chopped mode with a frequency of about 70Hz and a duty cycle of 50%. Thus the mode of operation can be regarded as quasi cw.

The tube of the FIR laser is a 1.5m long Pyrex waveguide with an inner diameter of 35mm. The Fabry Pérot resonator consists of a hybrid mesh output coupler and gold coated copper end mirror. The CO_2 laser beam is coupled into the resonator through a central hole of 1.5mm in the end mirror. The hybrid coupler is a z-cut quartz plate, 3mm thick, covered by a dielectric coating in order to reflect not absorbed CO_2 radiation. A capacitive gold mesh with 500lpi was evaporated onto this coating. This coupler was not optimized to yield highest possible output on the 184.3μm line. Thus the maximum output over all wavelengths is about 90mW measured with an Scientech 360001 detector at 56W pump power.

The output coupler is mounted on a translation stage for tuning of the resonator length.

The wavelengths were measured by an external Fabry Pérot interferometer. A monochromator preselected a line before entering the Fabry Pérot. All wavelengths were measured in air and the precision of the measurements was 0.05μm. The known wavelength of the 184.3μm line was taken as a reference from [4].

Pyroelectric detectors were positioned behind the monochromator and behind the Fabry Pérot interferometer. Their signals were monitored with a lock-in amplifier while the cavity lengths of the external Fabry Pérot interferometer and of the laser resonator were tuned.

Results and discussion

The results of the measurements are summarized in Table I.

The dominant lines in all experiments have been the 184.3μm, 195.84μm and 235.58μm lines. Optimizing gas pressure and resonator length 70mW, 32mW and 17mW, respectively, have been obtained at a pump power of 56W. Operating the two strongest lines simultaneously at 20Pa a total power of 90mW was achieved. From the ratio of the detector signal directly behind the monochromator the output power at 195.84μm is estimated to 20mW, that is 28% of the total power.

Table I:
Measured and calculated wavelengths of the laser lines pumped with the 9R32 CO_2 laser line, their polarisation and their optimum operating gas pressure;
* reference wavelength; † old line

λ/μm	transition	λ_{calc}/μm	pol.	opt. pressure/Pa	max. power/mW
	ν_9				
184.3 *†	$18_{17} \rightarrow 17_{16}$	184.19	⊥	20	70
195.84 †	$17_{16} \rightarrow 16_{15}$	195.70	⊥	19	32
208.83	$16_{15} \rightarrow 15_{14}$	208.76	⊥	13	12
223.99	$15_{14} \rightarrow 14_{13}$	223.72	⊥	8	weak
235.58 †	$18_{17} \rightarrow 18_{16}$	235.51	∥	20	17
250.61	$17_{16} \rightarrow 17_{15}$	250.45	∥	10	1.5
	ν_0				
172.50	$19_{18} \rightarrow 18_{17}$	172.54	⊥	9	weak
220.44	$18_{18} \rightarrow 18_{17}$	220.50	∥	12	1

The power level of the new 208.83μm line was estimated in the same way to yield a maximum of 12mW at 12Pa. The other two perpendicular polarized lines at 172.50μm and 223.99μm have been too weak to allow us measure their power on the strong background of the dominant lines.

The parallel polarized lines could be separated much easier. Scanning the cavity length their interferograms have much sharper maxima than the interferograms of the perpendicular polarized lines. Therefore it was possible to keep the laser running on only a single parallel polarized line. For the new lines at 220.44μm and 250.61μm power levels of 1mW and 1.5mW, respectively, have been obtained when the gas pressure was optimized.

For the perpendicular polarized lines the optimum gas pressure decreases with increasing wavelength. The 172.50μm line is an exception as is the 220.44μm line for the parallel polarized lines, whose optimum gas pressures also decrease with increasing wavelengths.

The power of each observed lines is more or less strongly oscillating at some resonator lengths in phase with one or more other lines. Operation of the 223.99μm, 208.83μm or 250.61μm decreases the power of the 235.58μm line. The stronger the 208.83μm, 195.84μm and 184.3μm lines are operating the better worked the 223.99μm line. These are typical properties of cascade transitions.

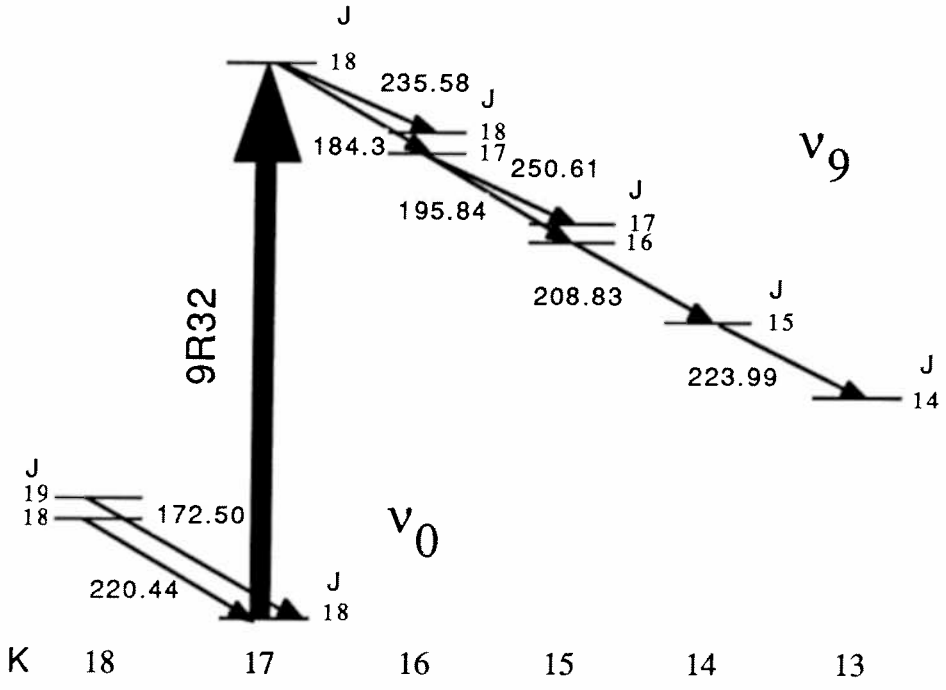
The assignment of the laser lines to rotational transitions was made using the effective Hamiltonian of Hirota and Sahara [5] and Hirota's measured molecular constants of $^{12}CH_2F_2$ [6]. His constants of the n_9 and n_3 states, which include the coriolis interaction between these two states, were used here. In this case the centrifugal constants were fixed to the ground state values. Table II contains the used data.

If the absorption of the 9R32 CO_2 line is regarded as a transition ($18_{17} \rightarrow 18_{17}$) [4,2], four lines with perpendicular polarisation can be assigned to the first steps of the $\Delta J=\Delta K=-1$ cascade starting at (J=18, K=17) as it was already proposed for the first two lines [4,2]. Two parallel polarized lines are assigned to the $\Delta J=0$, $\Delta K=-1$ transitions in the first two steps of this cascade. The 235.58μm line was already assigned. [4] Calculated and measured wavelengths agree quite well if one takes into account that the molecular data were derived from measurements of transitions with low J values. [6]

Table II:
Molecular constants of $^{12}CH_2F_2$ for the ground state and the ν_3 and ν_9 states in MHz; data in brackets are fixed to ground state values. [6]

	ν_0 (ground state)	ν_3	ν_9
$\tilde{A}$	49 142.818	48 295.44	48 704.26
$\tilde{B}$	10 604.7050	10 544.10	10 524.25
$\tilde{C}$	9 249.8437		
$\tilde{C}^*$		9 261.2	9 147.3
τ_{aaaa}	-2.30061	[-2.30061]	[-2.30061]
τ_{bbbb}	-0.06151	[-0.06151]	[-0.06151]
τ_{cccc}	-0.02723	[-0.02723]	[-0.02723]
T_1	0.1575	[0.1575]	[0.1575]
T_2	-0.03756	[-0.03756]	[-0.03756]
D		10 189	
F		-116	
ν_3-ν_9		628 300	

Figure 1:
Rotational energy levels of the vibrational ν_0 (ground) state and the ν_9 band of $^{12}CH_2F_2$ as well as the measured transition wavelengths in µm



The 172.50μm line and the 220.44μm line are assigned to refill transitions in the vibrational ground state. The (J=18, K=17) state in the vibrational ground state is strongly depleted by the pump laser so that in the (J=18, K=18) and (J=19, K=18) levels overpopulation is achieved. The polarisation rules remain the same as for the transitions in the excited vibrational state. Thus these lines can be attributed to the transitions ($19_{18} \rightarrow 18_{17}$) and ($18_{18} \rightarrow 18_{17}$) in the vibrational ground state. Measured and calculated wavelengths agree in the order of magnitude of the accuracy of the measurements.

Figure 1 shows some rotational energy levels of the vibrational ground state and the ν_0 band of $^{12}CH_2F_2$ as well as the assignment of the observed lines.

The power ratios of the FIR lines as well as the shift of optimum gas pressure can easily be understood by the assignement to a cascade. The relaxation rate γ_{V-t} was found to be very fast [4]. Decreasing the pressure means reducing the vibrational-translational and the rotational relaxation due to molecular collisions, which are important processes for the depopulation of the levels involved in the laser transitions at the bottom of the cascade. Reducing the collision rates improves the laser action for these transitions as well as for the refill transitions in the vibrational ground state.

Summary

A set of three new FIR laser lines of $^{12}CH_2F_2$ pumped by the 9R32 CO_2 laser line could be assigned to cascade transitions in the ν_9 vibrational level. Another pair of laser lines was discovered and has been assigned to refill rotational transitions in the vibrational ground state. Calculated and measured wavelengths agree very well.

Acknowledgements

The author wishes to thank Dr.R.Behn and Dr.M.R.Siegrist for fruitful discussion of the transition assignments.

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