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IN CH₃F**

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Abstract

A new laser line of CH₃F at 676 μ m has been found. The gas was pumped by a powerful pulsed CO₂ laser at 9P20. The line could be observed simultaneously with the 496 μ m line and can be assigned to a rotational transition in the 2 ν_3 band which is stepwise pumped from the vibrational ground state via the $\nu_3=1$ state.

Introduction

The optical pumping of CH₃F with a CO₂ laser at 9P20 usually activates the strong laser line at 496 μ m and some other cascade transitions [1,2]. Our CO₂ laser system provides pulses with a length of about 2 μ s and a peak power of 250-500MW. Under these very special pumping conditions a new laser line at 676 μ m could be observed in CH₃F which was even stronger than the 496 μ m line. We investigated the influence of the CH₃F pressure as well as the power and polarization of the pump laser on the output of the FIR laser. It was possible to identify the new line as the J=9 $\rightarrow$ 8 transition in the 2 ν_3 vibrational band. The excitation mechanism of the line is discussed.

Experiment

The CO₂ pump laser system consists of a hybrid TEA oscillator and a triple pass amplifier stage operating at 2 bar. The oscillator produces up to 80mJ single mode pulses for strong CO₂ lines. They are amplified at the 9P20 line up to 500J and have a duration of up to 2 μ s. With the resulting peak power of 500MW a pump intensity of up to 1.6MW/cm² is achieved in the 20cm diameter FIR resonator. Figure 1 shows the temporal shape of a typical CO₂ pump pulse. The repetition rate of the laser system is about one shot every two minutes. Therefore the number of measurements with this system is limited.

FIGURE 1

A typical CO laser pulse

The FIR laser was originally optimized for D₂O laser operation at 385 μ m [3]. Its unstable resonator configuration is formed by a concave spherical mirror ($f=5.15$ m, $\varnothing=200$ mm) and a convex spherical mirror ($f=-1.15$ m, $\varnothing=42$ mm). Therefore from the geometric point of view only 5% of the beam is reflected back into the resonator by the small mirror.

The resonator is L-shaped and the pump laser is coupled into the resonator through a free standing wire grid (tungsten, $a=10\mu$ m, $g=100\mu$ m). The transmission of this grid for CO₂ laser radiation is high and nearly independent of the polarization. However it fully determines the polarization of the FIR laser output. For wavelengths longer than 400 μ m the reflection is more than 98% for the polarization parallel to the wires and less than 10% for the polarization perpendicular to the wires [4,5]. Therefore all laser lines obtained with this laser are polarized parallel to the wires of the coupling grid.

A mylar foil reflects a small part of the laser beam onto a fast pyroelectric detector (Molelectron P5-01). A highpass waveguide filter [6] is placed in front of this detector to suppress radiation with wavelengths longer than 510 μ m. The main part of the beam is focussed onto the entrance slit of a 0.25m Ebert monochromator. A fast hot electron Ge-detector IPA 300F2 from the A.F.IOFFE Physico-Techninal Institut in St.Petersburg measures the power transmitted through the monochromator. Pulse energies can be measured by inserting a power meter PM103 from Thomas Keating Ltd..

Measurements

By pumping CH₃F at several Torr with the 9P20 line of our relatively long pulsed CO₂ laser we observed a new strong laser line at 676 μ m. The

wavelength was determined by means of the monochromator with a precision of approximately $\pm 2 \mu\text{m}$.

The appearance of the new line was independent of the polarization of the pump beam while the prominent line at $496 \mu\text{m}$ was only observed by pumping with a beam polarized perpendicular to the FIR laser polarization prescribed by the FIR resonator.

The known refill and cascade transitions at $419 \mu\text{m}$, $452 \mu\text{m}$, $541 \mu\text{m}$, $595 \mu\text{m}$ were not observable.

By pumping with a 320J (appr. 300MW peak power) CO_2 laser pulse polarized perpendicular to the FIR polarization a total FIR output of 45mJ was obtained at 3.5Torr CH_3F pressure. 8mJ can be attributed to the $496 \mu\text{m}$ line and 37mJ to the $676 \mu\text{m}$ line. These values are relatively low compared to the energy of the pump pulse. An enhancement of the FIR pulse energy at $496 \mu\text{m}$ should be possible by optimizing the FIR resonator parameters and by increasing the pump intensity. The threshold for the $496 \mu\text{m}$ line was determined by Biron et al. [7] in an ASE laser to be $0.7 \text{MW}/\text{cm}^2$ and for a superradiant laser by Evans et al. [8] to be $4 \text{MW}/\text{cm}^2$. Furthermore the rise time of our pump pulse of approximately $1 \mu\text{s}$ (figure 1) is long compared to the collisional thermalisation time for the rotational levels in the upper vibrational state which is about 2ns for 4.5Torr CH_3F pressure [9]. This might be another reason for the low laser output at $496 \mu\text{m}$.

The new line always shows a sharp onset and a fast rise in intensity. The pulse length is about $1 \mu\text{s}$ and decreases with increasing gas pressure. Figure 2 shows the FIR pulses at both lines from the same shot.

FIGURE 2

FIR emission at 496 μm and 676 μm for the same laser shot (no relative scaling)

The dependence of the FIR pulse energy at $676\mu\text{m}$ on the gas pressure is shown for both pump polarizations in Figure 3. The maximum value is obtained at a pressure of 4.5 Torr. For the pump pulse polarized parallel to the FIR laser polarization the efficiency of the FIR laser is obviously higher.

FIGURE 3

Pressure dependence of the FIR emission

The onset of the new line is delayed relatively to the beginning of the CO₂ laser pulse and to the pulse of the 496 μ m line. The delay decreases with the pump energy (Figure 4).

FIGURE 4

Delay of the 676 mm emission relative to the pump pulse

Line assignment and excitation mechanism

In Figure 5 an assignment for the new line is proposed.

Using the molecular constants of CH_3F derived by Freund et al. [10] the main absorption of the 9P20 pump line can be assigned to the Q(12,K) transition in the $\nu_3=1$ band [1]. This transition pumps the $496\mu\text{m}$ line which can be observed as Raman-like or as a center line transition.

The transition P(2,K) can also contribute to the population of the $\nu_3=1$ levels. The corresponding rotational transition in the $\nu_3=1$ band from $J=1$ to $J=0$ would have a wavelength of $5949\mu\text{m}$ but has never been reported as a laser transition. The detuning of this pump transition from the pump laser line is approximately 325MHz.

The fast collisional transfer between the rotational levels ($\tau_R=2\text{ns}$ for 4.5 Torr) [9] populates the $J=8$ level, and the $J=9$ level in the $\nu_3=2$ band can now be pumped by the R(8,K) transition. The detuning of this pump transition from the 9P20 line of 278MHz for $K=0$ can be compensated by the pressure broadening in CH_3F of 40MHz/Torr [9] and the tunability of the pump laser (100MHz).

The new laser line can now be attributed to the transition $J=9 \rightarrow J=8$ in the $\nu_3=2$ band. The calculated wavelength for $K=0$ is $669.7\mu\text{m}$. This deviation from the experimental value is not surprising since the molecular data were derived from measurements at levels with small J [10]. It cannot be fully excluded that the refill transition $J=9$ to $J=8$ in the $\nu_3=1$ state is the new laser transition. However the calculated wavelength for this transition is 661.2mm and deviates much more from the measurement than the calculated wavelength for the transition in the $\nu_3=2$ state.

FIGURE 5

Partial level diagram of the v_3 excited vibrational states of CH_3F with pump and laser transitions

The polarisation behaviour indicates that the new line observed at $676 \mu\text{m}$ is probably a real laser transition. In a Raman transition the polarization of the FIR beam would be linked to that of the pump beam which is in fact the case for the $496 \mu\text{m}$ line. However the new line appears on both pump polarizations. A slight preference for pumping parallel polarized to the FIR polarization was observed which is consistent with the line assignment.

Obviously the molecules are excited to a real upper laser level and lose information about the pump beam polarization by phase changing or K-changing collisions.

The fact that the new line has never been observed before can easily be explained. The comparably high energy of our pump pulse is sufficient to transfer a large number of molecules to the $v_3=1$ state. Furthermore the pump pulse has a duration of the order of the relaxation time τ_{vv} between the vibrational levels ($2\mu s$) [9]. This means that molecules which are pumped once into the $v_3=1$ state remain there during the pumping. The long duration of the pump pulse allows for a complete thermalisation in the $v_3=1$ state and therefore a sufficient population of the lower level for the R(8,K) pump transition. On the other hand the long rise time of the pump pulse however reduces the efficiency for $496\mu m$ operation.

Summary

A new laser line has been observed in CH_3F pumped by a pulsed CO_2 laser on the 9P20 line. It is assigned to the $J=9 \rightarrow J=8$ transition in the $v_3=2$ band. The upper laser level is pumped via the Q(12,K) and P(2,K) transitions from $v=0$ to $v_3=1$ and the R(8,K) transition from $v_3=1$ to $v_3=2$. The appearance of the line in our laser system results from our very special pumping conditions. By improving the FIR resonator for this line an even higher output could probably be obtained.

Acknowledgements

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References

- [1] T.Y.Chang and T.J.Bridges, *Opt. Comm.* **1**, 423 (1970).
- [2] F.Brown, S.Kronheim and E.Silver, *Appl. Phys. Lett.* **25**, 394 (1975).
- [3] S.A.Salito, Thesis, Ecole Polytechnique Fédérale de Lausanne (1989).
- [4] A.Blanco, S.Fonti and A.Piacente, *Infrared Phys.* **26**, 357 (1986).
- [5] E.A.Lewis and J.P.Casey, Jr., *J. Appl. Phys.* **23**, 605 (1952).
- [6] F.Keilmann, *Int. J. Infrared Millimeter Waves* **2**, 259 (1981).
- [7] D.G.Biron, B.G.Danly, R.J.Tempkin and B.Lax, *IEEE J. Quant. Electron.* **QE-17**, 2146 (1981).
- [8] D.E.Evans and L.E.Sharp, *Appl. Phys. Lett.* **26**, 630 (1975).
- [9] A.T.Rosenberger and T.A.DeTemple, *Phys. Rev. A* **24**, 868 (1981).
- [10] S.M.Freund, G.Duxbury, M.Römheld, J.T.Tiedje and T.Oka, *J. Mol. Spectrosc.* **52**, 38 (1974).