

FAR INFRARED LASING FREQUENCIES OF CH₂DOD*

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Abstract

We obtained laser action from the CH₂DOD molecule optically pumped by CO₂ laser radiation. Eight lasing transitions were identified as originating from CH₂DOD; an additional 21 transitions also lased, but were assigned to the CH₂DOH molecule, and 2 to CH₃OH, even though the isotopic purity of the sample was given as 98%. The relative intensity, relative polarization and frequency of all the lines were measured. The eight lines are distributed between 145.8 μm and 479.2 μm.

Normal methyl alcohol, CH₃OH, is the best known optically pumped FIR lasing molecule. The ¹³C and D isotopic species still have their vibrational absorption spectra in the CO₂ laser frequency range and hence are also good lasing gases. The isotopic variety CH₂DOH has been pumped by a CO₂ laser and shown to lase by Ziegler and Dürr (1) and subsequently by Scalabrin et al.(2) who

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found new lines and measured the frequencies of many lines. These data are condensed in a review paper by Kon et al.(3), and are listed in a review of frequency measured FIR lines (4).

In this work we report the irradiation of the CH₂DOD molecule with CO₂ laser radiation, obtaining laser action on 31 lines, from which just 8 appear to originate from transitions occurring in the CH₂DOD molecule. The other 23 lines were assigned as follows: 21 have been found previously in CH₂DOH(1,2) and 2 are known lines in CH₃OH. The wavelengths of the 8 new lines were obtained in a 98% pure sample of CH₂DOD, and range from 145.8 μm to 479.2 μm . They were obtained in a transversely pumped 1 m Fabry-Perot resonator with a conventional CO₂ pump laser as described previously (5).

We present, in Table I, the observed CH₂DOD laser lines along with their relative polarization, optimum pressure, relative intensity, and pump power. Two of the strongest lines, $\lambda=368.0 \mu\text{m}$ and $\lambda=413.1 \mu\text{m}$ were pumped by the 9P(32) CO₂ laser line. On this same pump line, Dyubko et al. (6) reported in CH₃OH two FIR lines at wavelengths $(372 \pm 1) \mu\text{m}$ and $(418 \pm 1) \mu\text{m}$ but these have not been found in our laser using a sample of CH₃OH (7); therefore we think they are actually due to CH₂DOD.

Table II is a summary of the CH₂DOD laser frequency measurements. We have measured all the frequencies of the 31 lines we have observed in our sample and then discovered that 21 of these correspond to the frequencies of the lines already measured in CH₂DOH. The measurements were in agreement ($\Delta\nu/\nu$) within a 1σ uncertainty of 3×10^{-7} . This uncertainty is mainly due to the reproducibility in setting this transversely pumped FIR laser two independent times; thus, there is a single-time frequency resettability of $1/\sqrt{2}$ times 3×10^{-7} , ie. 2×10^{-7} . Our early measurement made in a longitudinally pumped laser exhibited an uncertainty of $\pm 5 \times 10^{-7}$, and we think that this number represents the uncertainty in resettability in longitudinally pumped lasers due to feedback in the CO₂ laser.

Before the frequency measurements, wavelength measurements were made with $\pm 0.1 \mu\text{m}$ accuracy by counting the modes in a calibrated 5 mm scan of one end mirror of

the Fabry-Perot resonator. The polarizations of the FIR lines relative to the CO₂ lines were determined by using a metal-mesh or stacked-plate polarizer in front of the FIR detector. Optimum lasing pressures were measured with a thermocouple gauge, with the laser adjusted to obtain maximum FIR output intensity. Output intensities were measured with either a Golay cell or a metal-insulator-metal, point-contact diode.

During the measurements, we needed a continuous flux of the CH₂DOD vapor in order to maintain laser oscillation, apparently due to a rapid isotopic rearrangement of the deuterium and hydrogen in the CH₂DOD. The lines which increase with the flow rate of CH₂DOD are those attributed to CH₂DOD, and those which decrease with flow of CH₂DOD were attributed to CH₂DOH. Wall preparation, performed by filling the laser with CD₃OD several times for many minutes before the addition of CH₂DOD, did not seem to change the results.

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TABLE I - SUMMARY OF CH_2DOD LASER LINES

CH_2DOD Laser Line λ (μm)	Rel. Pol.	CH_2DOD Press. Pa(mTorr) ^(a)	Relative FIR Int.	CO_2 Power (W)	CO_2 Pump Line
145.8		24(180)	6	33	10P(30)
158.4	⊥	40(300)	100	41	10P(10)
267.7		13(100)	10	36	10R(16)
278.9	⊥	20(150)	8	36	10R(16)
479.2		20(150)	18	36	10R(16)
368.0(372±1) ^(b)	⊥	20(150)	90	24	9P(32)
413.1(418±1) ^(b)		17(130)	60	24	9P(32)
230.6	⊥	27(200)	12	18	9P(06)

^(a) 1 Torr = 133.3 Pa.

^(b) Reported by Dyubko et al. (1981) in CH_3OH , but not found in our laser with a sample of $\text{CH}_3\text{OH}^{(5)}$.

TABLE II - SUMMARY OF CH₂DOD LASER LINE FREQUENCY MEASUREMENTS

CH ₂ DOD Laser Line λ (μm)	Measured Freq. (MHz) Uncertainty $\Delta\nu/\nu = 2 \times 10^{-7}$	Vacuum Wavenumber (cm^{-1})	CH ₂ DOD Press. Pa(mTorr)	CO ₂ Pump Line
145.769 789	2 056 615.851	68.601 321	24(180)	10P(30)
158.390 159	1 892 746.748	63.135 236	27(200)	10P(10)
230.610 467	1 299 995.010	43.363 166	27(200)	9P(06)
267.660 699	1 120 046.608	37.360 733	13(100)	10R(16)
278.930 573	1 074 792.392	35.851 215	21(160)	10R(16)
367.974 974	814 708.824	27.175 761	20(150)	9P(32)
413.113 483	725 690.326	24.206 424	20(150)	9P(32)
479.226 106	625 576.224	20.866 977	20(150)	10R(16)