Heterodyne Frequency Measurements and Analysis of CO_2 Laser Hot Band Transitions

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New frequency measurements of molecular lines in the P branch of the 01^11-$[11^10, 03^10]_I$ band of $^{12}C^{16}O_2$, observed in laser emission, and lines in the R branch, observed in absorption with a tunable-diode laser, have been made by heterodyne difference frequency techniques, with stabilized CO_2 lasers generating the reference frequencies. The data obtained, plus additional results of measurements made by two other groups of researchers, were combined in a least-squares fit to obtain improved rovibrational constants for this band. The new constants predict more reliably the frequencies of both the P- and R-branch transitions. A table of transition frequencies and their estimated uncertainties is given for this band.

INTRODUCTION

The CO_2 laser, with its many isotopic variations, has become extremely useful as a secondary frequency standard in the infrared (*1*). Although most of the measurements and analyses have involved the main laser transitions (the 00^01-$[10^00, 02^00]_I$ and 00^01-$[10^00, 02^00]_{II}$ bands), other higher-level transitions have been successfully made to lase and can be used as frequency standards as well, provided the frequencies are accurately known (*2*). One such set of higher-level laser transitions belongs to the 01^11-$[11^10, 03^10]_I$ hot band.

The frequencies of some transitions of the CO_2 hot band have previously been measured quite accurately by Whitford *et al.* (*3*). This earlier work involved P-branch measurements, one measurement of a Q-branch transition, and an R-branch transition. With nearly the same accuracy, the wavelengths (wavenumbers) of many P-branch transitions have been measured by Monchalin *et al.* (*4*).

In this paper we report some new measurements made on these transitions and an analysis that includes the earlier measurements. The present measurements are

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intended (a) to provide yet another independent set of frequency measurements, (b) to provide better coverage of both the *P*- and *R*-branch transitions, and (c) to provide an improved table of frequencies and their uncertainties for these transitions.

EXPERIMENTAL DETAILS

The frequencies of 12 (new) *P*-branch transitions were measured at the National Research Council of Canada and are listed in Table I. The laser lines from $P(43)$ to $P(33)$ and $P(9)$ were centered to the peak power and heterodyned against two reference lasers. The two reference frequencies were stabilized by saturated absorption (5) and selected from the well-known 9- and 10-μm CO_2 laser bands so that $2\nu_2 - \nu_1 \cong \nu_3$, where ν_3 is the frequency of the hotband transition to be measured. All three laser beams were focused onto a W–Ni point–contact diode. The diode produced a beat frequency which was measured with a spectrum analyzer and frequency counter (6). The main uncertainty in this measurement arises from setting the hot band laser line frequency to the peak power. Unfortunately, the 4.3-μm saturated fluorescence stabilization did not produce enough signal/noise to be used.

The *R*-branch measurements were made at the National Bureau of Standards by a TDL (tunable-diode laser) heterodyne technique (7, 8), in which the frequency of the TDL was locked to the absorption line center of the CO_2 hot band transition. The frequency-locked output was then heterodyned with a CO_2 reference laser, and the frequency difference was measured with a spectrum analyzer. Six hot band *R*-branch transitions between $R(4)$ and $R(52)$ were measured in this manner. The TDL power and 1-GHz, 3-dB bandwidth of the Hg–Cd–Te detector limited the number of lines on which measurements could be made. Observable beat frequencies (between the CO_2 laser reference and the hot band-referenced TDL) of slightly over 3.1 GHz permitted six measurements. The specific lines measured in both branches are listed in Table 1.

Since it was possible to actively stabilize the NBS CO_2 hot band laser to the saturated absorption feature, nine lines approximately equally spaced over the lasing range of the laser were measured. The experimental setup was similar to that used in Ref. (2), except that only one reference laser stabilized by saturated absorption was used. The difference frequency between the hot band and reference lasers was generated in the W–Ni point–contact diode by an appropriate harmonic combination of an 8- to 12- and a 58- to 65-GHz klystron. The beat signal, along with frequency markers, were averaged for several minutes on a signal averager. Typically, 10 runs were averaged together to produce a given line frequency measurement. The relative phase and amplitude of the dither voltages which modulated the laser frequencies were adjusted to minimize the width of the beat signal, typically about 50 kHz as observed on a spectrum analyzer. The room-temperature absorption cells used to stabilize the reference and hot band lasers were both filled to a pressure of 5.3 Pa with CO_2. The high-frequency klystron was phase locked to an X-band klystron; both X-band klystrons were phase locked to quartz crystals. The frequencies of the X-band klystrons were counted directly with counters using external standards traceable to the NBS Cs primary frequency standard. The results of the nine measurements are given in Table I.

TABLE I

TABLE I

Measured $^{12}C^{16}O_2$ Hot Band 01^11-[11^10, 03^10] Frequencies

J	$^{12}C^{16}O_2$ Ref. Freq.	$\nu_{Hot}-\nu_{Ref.}$[a] (MHz)	Measured Freq. (MHz)	Obs.-Calc[b] (MHz)
		Doppler Limited Measurements(NRC)[c]		
P-branch				
43	$2R_I(28)-R_{II}(12)$	1322.01	26 636 675.00 (60)	-0.94
42	$2P_I(36)-R_I(10)$	960.56	26 647 450.85 (60)	-0.03
41	$2R_I(32)-R_{II}(16)$	- 490.93	26 697 509.31 (60)	0.23
40	$2R_I(16)-P_{II}(10)$	333.25	26 709 734.71 (60)	0.36
39	$2P_I(30)-R_{II}(22)$	1067.84	26 757 659.53 (60)	-0.09
38	$2R_I(40)-R_{II}(26)$	-1453.32	26 771 244.65 (60)	0.12
37	$2R_I(38)-R_{II}(20)$	627.25	26 817 128.96 (60)	-0.04
36	$2R_I(16)-P_{II}(30)$	1437.60	26 831 984.34 (60)	0.27
35	$2P_I(30)-R_I(16)$	489.69	26 875 918.37 (60)	-0.16
34	$2R_I(10)-P_I(26)$	- 571.30	26 891 955.39 (60)	-0.01
33	$2R_I(38)-R_{II}(14)$	1969.55	26 934 029.43 (60)	0.02
9	$2P_I(22)-R_I(16)$	2134.20	27 578 702.70 (60)	-0.61
		Saturated Absorption Measurements(NRC)[d]		
P-branch				
29	$P_I(50)$	-365 381.12	27 048 219.29 (20)	-0.07
24	$P_I(50)$	-233 238.23	27 180 362.18 (20)	-0.13
23	$P_I(56)$	41.69	27 214 437.86 (20)	0.32
22	$P_I(50)$	-177 834.61	27 235 765.80 (20)	-0.09
20	$P_I(50)$	-123 187.15	27 290 413.26 (20)	-0.16
19	$P_I(50)$	- 91 721.49	27 321 878.92 (20)	-0.17
17	$P_I(42)$	-293 684.62	27 374 589.83 (20)	-0.08
16	$P_I(50)$	- 16 156.35	27 397 444.06 (20)	-0.27
15	$P_I(50)$	13 027.19	27 426 627.60 (20)	-0.11
14	$P_I(48)$	- 28 601.19	27 449 828.96 (20)	-0.30
12	$P_I(50)$	87 860.70	27 501 461.11 (20)	-0.17
11	$P_I(50)$	115 083.99	27 528 684.40 (20)	-0.09
Q-branch				
11	$P_I(38)$	-4348.50	27 786 661.90 (20)	0.16
R-branch				
23	$P_I(20)$	47.96	28 306 272.85 (20)	-0.12
		Doppler Limited Measurements (NBS)[e]		
R-branch				
4	$P_I(34)$	- 781.8	27 909 939.0 (40)	-0.3
12	$P_I(28)$	-1390.4	28 083 279.4 (40)	-0.7
26	$P_I(18)$	-2590.1	28 357 183.7(100)	-3.2
32	$P_I(14)$	-1738.6	28 462 935.1 (50)	-3.4
47	$P_I(4)$	3102.9	28 717 240.6 (50)	-1.6
52	$P_I(2)$	1684.3	28 763 525.2 (50)	-3.7
		Saturated Absorption Measurements (NBS)[f]		
P-branch				
35	$P_I(50)$	-537 681.8769	26 875 918.5346(150)	0.0005
32	$P_I(50)$	-462 439.6017	26 951 160.8098(150)	-0.0028
29	$P_I(50)$	-365 381.0460	27 048 219.3655(150)	0.0017
26	$P_I(50)$	-289 398.9282	27 124 201.4833(150)	-0.0068
23	$P_I(50)$	-199 162.8678	27 214 437.5437(150)	0.0051
20	$P_I(50)$	-123 186.9829	27 290 413.4286(150)	0.0109
18	$P_I(44)$	-261 456.6380	27 344 305.9459(150)	0.0023
15	$P_I(50)$	13 027.2872	27 426 627.6987(150)	-0.0119
12	$P_I(50)$	87 860.8718	27 501 461.2833(150)	0.0074

a) The estimated uncertainty in the last digits is listed in parentheses.
b) The calculated values in Table III were obtained by using the constants from Table II.
c) P(43) to P(33) and P(9) are new measurements made at NRC on lines observed in laser emission.
d) Measurements made earlier at NRC, but corrected to reflect new values[1] for the CO_2 reference lasers.
e) The Doppler limited R-branch lines were measured by tunable diode laser heterodyne techniques.
f) New measurements made at NBS. Each laser frequency was actively stabilized to the saturated absorption line with a room temperature absorption cell filled to 5.3 Pa pressure with CO_2.

ANALYSIS OF DATA AND CALCULATION OF TRANSITION FREQUENCIES

In addition to the earlier measurements of the 01^11-$[11^10, 03^10]_1$ laser transitions (3, 4), there are measurements of the absorption spectrum of CO_2 that are valuable to the present analysis. Bailly et al. (9) have used Fourier transform spectroscopy (FTS) to measure transitions involving the upper energy levels of the laser hot band transitions.

In order to obtain the best parameters for calculating the energy levels and transition frequencies, a least-squares fit was made which combined the present measurements with the measurements given in Refs. (3), (4), and (9). However, the actual frequencies given in Ref. (3) were not used. Instead, we recalculated the frequencies from the raw experimental measurements using the latest values for the CO_2 laser frequencies (1). These recalculated frequencies are also given in Table I. Also, the measured wavelengths in Ref. (4) were adjusted to reflect the present recommended value for the wavelength of $^{127}I_2$, transition 11-5 $R(127)$, component i (632 991 398.1 fm ± 1 part in 10^9) (10), which was the reference standard used in that experiment. The least-squares fits were made by using the same double-precision computer program as was used by Petersen et al. (2) for the $^{13}CO_2$ hot band laser transitions.

In the present analysis, each measurement was weighted by the inverse square of the estimated uncertainty of the measurement. For the data resulting from the work of Ref. (3), we used an estimated uncertainty of 0.2 MHz, and the data from Ref. (4) were given an uncertainty of 0.3 MHz. The data of Ref. (9) were weighted as indicated in that paper, which is equivalent to giving an uncertainty of 1.0 MHz to those data that have a weight of 1 in Ref. (9). Although we believe the FTS data have an absolute uncertainty greater than 1 MHz, only the relative accuracy between the FTS measurements will have any effect on the hot band laser frequency calculations, since the FTS measurements only contribute to determining the rotational constants. That is to say, the constants for the hot band laser transitions are only sensitive to the frequency differences between the lines measured by the Fourier transform technique rather than to the absolute frequency of each line.

The wavenumber data given in Ref. (4) and (9) were converted to frequency units by multiplying by the speed of light (c = 299 792 458 m/s). With this conversion the wavelength measurements of Monchalin et al. (4) are in almost perfect agreement with the present measurements and analysis. The largest deviation between the calculated transition frequencies and the frequencies obtained from the data of Monchalin et al. is 0.3 MHz.

The data were fit to the term value equations

$$F(v, J) = G_v + B_vJ(J + 1) - D_v[J(J + 1) - 1]^2$$
$$+ H_v[J(J + 1) - 1]^3 \pm 1/2J(J + 1)[q_v - \mu_vJ(J + 1)] \quad (1)$$

with

$$\nu_{obs} = F'(v', J') - F''(v'', J'') \quad (2)$$

and

$$\nu_o = G' - G'', \quad (3)$$

where the $'$ and $''$ indicate the upper and lower states, respectively; and we have used the symbol μ_v where some other papers use q_{vJ}. The last term of Eq. (1) takes into account the l-type doubling effect. For the levels with which this paper is concerned, the proper sign to use for the last term in Eq. (1) is as follows: since the upper state is a gerade (g) state, the upper sign ($+$) is used for the odd J values and the lower sign ($-$) is used for the even J values; the opposite signs are used for the lower ungerade (u) state. This sign convention gives q_v a positive value.

The constants resulting from this fit are given in Table II. As one would expect, the constants for the upper state are not very different from those given by Bailly *et al.* The constants for the laser transitions are also quite close to the values given in Ref. (4), although the present constants are much more accurate.

Since $H(01^11)$ is so small, it is not determined, but we have retained this constant in the fit so its uncertainty will be properly reflected in the uncertainty of the calculated laser frequencies. We were, however, able to obtain a good value for the difference in the H values, so we have also given in Table II the value for $H(01^11)$-$H[11^10, 03^10]_1$. Its value is the same order of magnitude as that found for the corresponding band of $^{13}CO_2$ in Ref. (2). The value of $H[11^10, 03^10]_1$ is over six times larger than the value of H_0 found by Bailly *et al.*, while the value of $H(01^11)$ is much closer to their value for H_0. This can be readily explained since the perturbation due to the Fermi resonance between the 03^10 and 11^10 states would be expected to give both D and H values that are somewhat different from the unperturbed values, and the unperturbed values would be expected to be very close to the ground state values.

The present value for the band center, ν_0, is only 1.5 MHz lower than that given by Monchalin *et al.* (4), within twice the ± 1 MHz uncertainty given by them.

The constants given in Table II were used to calculate the frequencies given in

TABLE II

Rovibrational Constants for the 01^11-$[11^10, 03^10]_1$ Band of $^{12}C^{16}O_2$

Constants	(MHz)[a]
$\nu_0(01^11$-$[11^10,03^10]_1)$	27 795 449.889(46)
$B(01^11)$	11 628.687 17(269)
$D(01^11)$	4.051 881(1925) $\times$ 10^{-3}
$H(01^11)$	5.84(3147) $\times$ 10^{-11}
$q(01^11)$	17.916 769(1003)
$\mu(01^11)$	2.7072(581) $\times$ 10^{-5}
$B[11^10,03^10]_I$	11 718.038 33(257)
$D[11^10,03^10]_I$	3.699 985(1694) $\times$ 10^{-3}
$H[11^10,03^10]_I$	1.9525(2568) $\times$ 10^{-9}
$q[11^10,03^10]_I$	27.708 361(898)
$\mu[11^10,03^10]_I$	-1.404 67(526) $\times$ 10^{-4}
$H(01^11)$-$H[11^10,03^10]_I$	-1.894 1(1323) $\times$ 10^{-9}

a) The uncertainty in the last digits (one estimated standard error) is given in parentheses.

TABLE III

Frequencies and Wavenumbers for the 01^{1}1-[11^{1}0, 03^{1}0]$_I$ Band of ^{12}C^{16}O$_2$
Calculated with the Constants from Table II

ROT. TRANS.	FREQUENCY (MHz)[a]	WAVENUMBER (cm^{-1})	ROT. TRANS.	FREQUENCY (MHz)	WAVENUMBER (cm^{-1})
P(55)	26257246.98(276)	875.847483(92)	R(0)	27818725.18(11)	927.932789(4)
P(54)	26257339.86(266)	875.850581(89)	R(1)	27841759.80(12)	928.701142(4)
P(53)	26322212.67(204)	878.014505(68)	R(2)	27864709.88(13)	929.466674(4)
P(52)	26324328.91(197)	878.085095(66)	R(3)	27887393.26(14)	930.223310(5)
P(51)	26386484.56(147)	880.158385(49)	R(4)	27909939.29(15)	930.975365(5)
P(50)	26390524.90(143)	880.293156(48)	R(5)	27932349.36(16)	931.722884(5)
P(49)	26450064.84(103)	882.279194(34)	R(6)	27954412.35(17)	932.458826(6)
P(48)	26455931.60(102)	882.474889(34)	R(7)	27976627.11(18)	933.199831(6)
P(47)	26512955.56(70)	884.377003(23)	R(8)	27998127.81(19)	933.917017(6)
P(46)	26520552.61(71)	884.630414(24)	R(9)	28020225.38(20)	934.654113(7)
P(45)	26575158.66(46)	886.451875(15)	R(10)	28041084.26(21)	935.349890(7)
P(44)	26584391.30(48)	886.759843(16)	R(11)	28063142.97(22)	936.085689(7)
P(43)	26636675.94(29)	888.503871(10)	R(12)	28083280.13(23)	936.757393(8)
P(42)	26647509.88(31)	888.863284(10)	R(13)	28105378.56(24)	937.494517(8)
P(41)	26697509.08(17)	890.533046(6)	R(14)	28124713.67(25)	938.139467(8)
P(40)	26709734.35(19)	890.940837(6)	R(15)	28146930.72(26)	938.880548(8)
P(39)	26757659.62(10)	892.539452(3)	R(16)	28165382.97(26)	939.496049(9)
P(38)	26771244.53(12)	892.992596(4)	R(17)	28187797.92(26)	940.243731(9)
P(37)	26817129.003(59)	894.523137(2)	R(18)	28205285.94(27)	940.827068(9)
P(36)	26831984.067(74)	895.018649(2)	R(19)	28227978.55(26)	941.584012(9)
P(35)	26875918.534(46)	896.484145(2)	R(20)	28244420.33(27)	942.132451(9)
P(34)	26891955.403(53)	897.019077(2)	R(21)	28267470.84(27)	942.901334(9)
P(33)	26934029.406(44)	898.422515(1)	R(22)	28282783.71(28)	943.412116(9)
P(32)	26951160.813(45)	898.993957(2)	R(23)	28306272.97(27)	944.195633(9)
P(31)	26991462.696(44)	900.338283(1)	R(24)	28320373.49(29)	944.665976(10)
P(30)	27009602.386(44)	900.943358(1)	R(25)	28344382.97(27)	945.466846(9)
P(29)	27048219.364(44)	902.231482(1)	R(26)	28357186.89(30)	945.893939(10)
P(28)	27067282.033(43)	902.867344(1)	R(27)	28381798.78(29)	946.714903(10)
P(27)	27104300.256(43)	904.102139(1)	R(28)	28393220.98(32)	947.095907(11)
P(26)	27124201.490(43)	904.765973(1)	R(29)	28418518.23(31)	947.939732(10)
P(25)	27159706.106(43)	905.950279(1)	R(30)	28428472.63(36)	948.271775(12)
P(24)	27180362.314(43)	906.639296(1)	R(31)	28454539.03(35)	949.141256(12)
P(23)	27214437.539(44)	907.775923(1)	R(32)	28462938.56(40)	949.421435(13)
P(22)	27235765.886(43)	908.487360(1)	R(33)	28489858.78(41)	950.319397(14)
P(21)	27268495.065(44)	909.579088(1)	R(34)	28496615.27(46)	950.544769(15)
P(20)	27290413.418(42)	910.310206(1)	R(35)	28524474.98(47)	951.474069(16)
P(19)	27321879.090(44)	911.359788(1)	R(36)	28529499.14(52)	951.641657(17)
P(18)	27344305.944(42)	912.107867(1)	R(37)	28558385.00(55)	952.605185(18)
P(17)	27374574.909(44)	913.118031(1)	R(38)	28561586.32(59)	952.711970(20)
P(16)	27397444.328(42)	913.880373(1)	R(39)	28591586.10(62)	953.712655(21)
P(15)	27426627.711(44)	914.853826(1)	R(40)	28592872.80(67)	953.755574(22)
P(14)	27449829.265(43)	915.627746(1)	R(41)	28624075.41(69)	954.796382(23)
P(13)	27477992.577(45)	916.567173(1)	R(42)	28623354.39(76)	954.772331(25)
P(12)	27501461.276(45)	917.350005(1)	R(43)	28655849.95(77)	955.856266(26)
P(11)	27528684.486(47)	918.258073(2)	R(44)	28653026.70(87)	955.762092(29)
P(10)	27552340.715(48)	919.047160(2)	R(45)	28686906.63(87)	956.892205(29)
P(9)	27578703.308(52)	919.926522(2)	R(46)	28681885.16(103)	956.724707(34)
P(8)	27602467.766(54)	920.719219(2)	R(47)	28717242.21(100)	957.904092(33)
P(7)	27628048.810(58)	921.572510(2)	R(48)	28709925.02(127)	957.660016(43)
P(6)	27651842.446(62)	922.366181(2)	R(49)	28746853.35(122)	958.891813(41)
P(5)	27676720.657(67)	923.196028(2)	R(50)	28737141.33(163)	958.567855(54)
P(4)	27700464.601(72)	923.988041(2)	R(51)	28775736.55(156)	959.855253(52)
P(3)	27724718.409(78)	924.797061(3)	R(52)	28763528.95(214)	959.448051(71)
P(2)	27748333.911(85)	925.584789(3)	R(53)	28803888.23(207)	960.794291(69)
P(1)	27772041.520(92)	926.375590(3)	R(54)	28789082.53(284)	960.300427(95)
			R(55)	28831304.62(278)	961.708804(93)

a) The estimated uncertainty (twice the standard error) in the last digits is given in parentheses following each value.

Table III for the 01^{1}1-[11^{1}0, 03^{1}0]$_I$ band of ^{12}CO$_2$. The wavenumbers given in Table III were calculated directly from the frequencies.

The uncertainties given in parentheses after the frequencies in Table III were estimated by using the elements of the variance–covariance matrix determined by the least-squares fit and given in Table IV. Since the measurements used in this analysis came from several sources and involved different measurement techniques, we believe that the uncertainties given in Table III accurately represent the uncertainties in the absolute frequencies of the calculated transitions.

TABLE IV

The Variance–Covariance Matrix (in units of MHz^2) for the Constants given in Table II

	ν_0	B'	D'	H'	q'	μ'	B''	D''	H''	q''	μ''
ν_0	2.11672×10^{-3}	1.05001×10^{-4}	6.80528×10^{-8}	1.02034×10^{-11}	-9.54486×10^{-7}	-3.65175×10^{-9}	1.03051×10^{-4}	6.29842×10^{-8}	9.03808×10^{-12}	-1.12304×10^{-6}	-3.46724×10^{-9}
B'		7.26024×10^{-6}	4.55277×10^{-9}	6.74080×10^{-13}	-2.33250×10^{-7}	-2.97537×10^{-10}	6.90096×10^{-6}	3.89689×10^{-9}	4.62571×10^{-13}	-2.24887×10^{-7}	-2.67860×10^{-10}
D'			3.70673×10^{-12}	5.90945×10^{-16}	-2.55602×10^{-10}	-2.09964×10^{-13}	4.36850×10^{-9}	3.23311×10^{-12}	4.30450×10^{-16}	-2.38112×10^{-10}	-1.83310×10^{-13}
H'				9.90165×10^{-20}	-4.42038×10^{-14}	-4.04919×10^{-17}	6.49010×10^{-16}	5.18330×10^{-16}	7.37324×10^{-20}	-4.13116×10^{-14}	-3.55307×10^{-17}
q'					1.00507×10^{-6}	1.98404×10^{-10}	-2.13600×10^{-7}	-2.03799×10^{-10}	-2.14125×10^{-14}	8.98802×10^{-7}	1.32539×10^{-10}
μ'						3.37825×10^{-13}	-2.80840×10^{-10}	-1.77001×10^{-13}	-2.75110×10^{-17}	1.91824×10^{-10}	3.03530×10^{-13}
B''							6.57997×10^{-6}	3.77082×10^{-9}	4.59472×10^{-13}	-2.07245×10^{-7}	-2.54197×10^{-10}
D''								2.87019×10^{-12}	4.00692×10^{-16}	-1.92160×10^{-10}	-1.57443×10^{-13}
H''									6.59412×10^{-20}	-2.14683×10^{-14}	-2.59473×10^{-17}
q''										8.06130×10^{-7}	1.33583×10^{-10}
μ''											2.76922×10^{-13}

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