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Heterodyne Frequency Measurements of Carbonyl Sulfide
Transitions at 26 and 51 THz. Improved OCS,
 $O^{13}CS$, and $OC^{34}S$ Molecular Constants

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Heterodyne frequency measurements were made on selected absorption features of carbonyl sulfide (OCS) near 26 THz (860 cm^{-1}) and 51 THz (1700 cm^{-1}). Frequency differences were measured between a tunable diode laser (TDL) locked to carbonyl sulfide absorption lines and either a stabilized $^{13}CO_2$ laser or a CO laser which was referred to stabilized CO_2 lasers. These measurements are combined with conventional TDL measurements and published microwave measurements to obtain new, more reliable molecular constants for OCS, $O^{13}CS$, and $OC^{34}S$. New frequency measurements are given for nine CO laser transitions between 1686 and 1726 cm^{-1} .

INTRODUCTION

The need for frequency calibration standards for the tunable diode laser (TDL) user community is well known and has frequently been brought to the attention of the National Bureau of Standards (NBS). Carbonyl sulfide (OCS) and nitrous oxide (N_2O) are two calibrants currently in use (1-4); NBS has on-going experiments to improve the accuracy of existing frequency tables for these and other molecules. The approach to obtain this improvement is to combine, in a least-squares fit, accurate heterodyne frequency measurements (TDL vs CO_2 laser, or TDL vs CO laser), published microwave measurements, and conventional TDL measurements made relative to existing absorption frequency standards of high quality. This procedure has decreased the uncertainty of $^{16}O^{12}C^{32}S$ frequencies between one and two orders of magnitude and is now being extended to the most abundant isotopic species of OCS.

One of the techniques being used to extend the calibration tables into spectral regions far from known laser transitions is to measure hot bands and apply the Ritz combination principle to obtain transition frequencies that cannot be directly measured. In Ref. (4), for example, the $20^{00}-00^{00}$ transitions for the $^{16}O^{12}C^{32}S$ isotopic species were determined by measuring the $20^{00}-10^{00}$ and the $10^{00}-00^{00}$ transition frequencies. The present work gives some direct measurements of the $20^{00}-00^{00}$ transitions near 51 THz and verifies our previous calculations for these frequencies and their uncertainties.

The primary motivation for this work, however, was to extend the range of accurate measurements on transitions for the $^{16}\text{O}^{12}\text{C}^{34}\text{S}$ and $^{16}\text{O}^{13}\text{C}^{32}\text{S}$ isotopic species which are present in a natural sample with abundances of 4.16 and 1.05%, respectively.

EXPERIMENTAL TECHNIQUE

The experimental technique consists of first locking a TDL to the carbonyl sulfide absorption feature of interest. A first-derivative locking technique is used in combination with an adjustment of a monochromator filtering the TDL radiation to produce a zero slope at the absorption line center. Another portion of the TDL beam is simultaneously heterodyned in a HgCdTe mixer with the beam from either a CO_2 or CO laser whose frequency is accurately known. The TDL-gas laser frequency difference is then added with appropriate sign to the gas laser reference frequency to give the OCS frequency value. Since the details of this technique have been described previously, (3-5) we restrict our explanation mainly to changes mandated by the use of the CO laser.

The 26-THz measurements of the 20^00-10^00 band were straightforward (4, 5) and were referenced to the $01^11-[11^10, 03^10]_1$ hot-band lasing transitions in the $^{13}\text{CO}_2$ laser (6). A HgCdTe heterodyne detector with a 1-GHz 3-db bandwidth near $10.4 \mu\text{m}$ is now part of our spectrometer. This new detector was used not only for the $11.8\text{-}\mu\text{m}$ measurements but also for the $5.9\text{-}\mu\text{m}$ work where this domestically manufactured device permitted the measurement of a 4.1-GHz frequency difference.

Even within the range of the fast HgCdTe mixer, there was little overlap of the CO laser frequency with the OC^{34}S and O^{13}CS spectra. Consequently, it was not possible to obtain the coverage in J that we desired in order to use the 20^00-10^00 measurements with previous 10^00-00^00 measurements to predict 20^00-00^00 transitions as accurately as we have done for normal OCS. In the current experiments, the lowest J values measured were 31 for OC^{34}S and 15 for O^{13}CS .

Subsequent to the 26-THz measurements, a $5.9\text{-}\mu\text{m}$ TDL which had a useable linewidth was used in a heterodyne system based on a CO laser, as shown in Fig. 1. Since the CO laser does not lend itself to a stabilization scheme of the required accuracy, it was necessary to synthesize a reference for the CO operating frequency. The apparatus for this synthesis included a MIM diode, two stabilized CO_2 lasers, and an X -band klystron (7).¹ The radiation from the CO laser, the two CO_2 lasers, and the klystron all impinged on the MIM diode. The beat frequency, ν_B , which propagated from the MIM diode was equal to

$$\nu_B = |\nu_{\text{CO}} - (3\nu_1 - \nu_2 \pm \nu_{\mu\text{w}})|, \quad (1)$$

where ν_{CO} , ν_1 , ν_2 , and $\nu_{\mu\text{w}}$ are the frequencies of the CO laser, a $^{13}\text{C}^{16}\text{O}_2$ laser, a $^{12}\text{C}^{16}\text{O}_2$ laser, and an X -band klystron, respectively. The beat signal, typically on the order of 100 MHz, was amplified and displayed on a spectrum analyzer. The CO laser was tuned to the midpoint between the two frequencies marking the disap-

¹ It has been demonstrated that with a MIM diode, two CO_2 lasers, and a klystron, one can synthesize and measure any frequency up to 150 THz. The power required from the unknown oscillator for an adequate S/N ratio of the beat signal varies from a few microwatts in the FIR to some tens of milliwatts at $2 \mu\text{m}$.

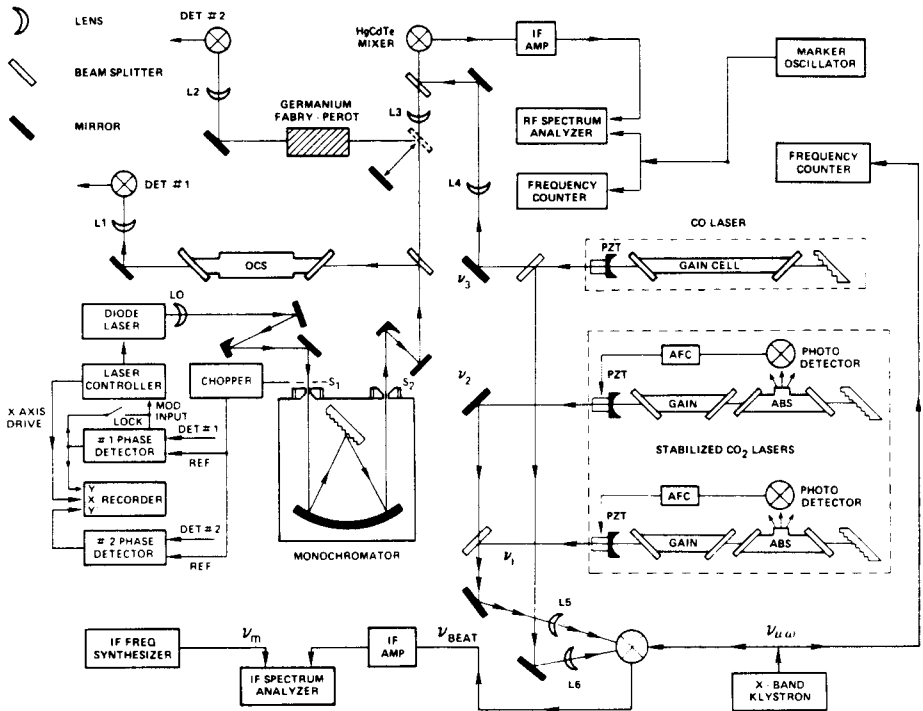


FIG. 1. Block diagram of a TDL-CO laser heterodyne spectrometer used for 51 THz measurements. The TDL was used in a closed cycle cooler for all of the measurements reported in this paper. The essential difference between this system and those we have described previously is in the use of CO₂ lasers to determine the frequency of the laser which is heterodyned with the TDL. The 1.25-m-long ¹²CO₂ laser was stabilized to an internal absorption cell; the ¹³CO₂ laser was 2 m long and stabilized to an external cell. Off-axis parabolas were again used as the reflecting optics at the input and output apertures of the monochromator. The OCS absorption cells were 1.7 m long. The heterodyne detector had a 1-GHz 3-db bandwidth. For the 26-THz operation, only the 2-m ¹³CO₂ laser was used (in the CO laser's diagrammed position), and it was then operated on the hot-band transitions.

pearance of the beat signal as the CO laser was tuned across its gain bandwidth. Thus, the measurement is also a determination of the CO laser line frequency. Table I summarizes these results along with the CO₂ synthesis combination used in the measurement. It should be emphasized, however, that the accuracy of the OCS measurement does not depend on locating the center of the CO lines since the CO laser frequency was manually adjusted to the measured value given in Table I while the TDL-CO laser frequency differences were measured.

We note that the CO frequency values in the literature were typically 20–30 MHz different from our measured values; hence our measurements were essential to achieving our objective of obtaining an accuracy of ± 3 MHz for the OCS frequencies. Even so, we did not always meet this objective for individual measurements because of the TDL linewidth, but the data allow us to calculate many OCS frequencies with the desired accuracy.

TABLE I
Frequency Synthesis Measurements of $^{12}\text{C}^{16}\text{O}$ Laser Lines

$^{12}\text{C}^{16}\text{O}$ Laser	ν_1^a	ν_2^a	ν_{CO}^b
Transition	$^{13}\text{C}^{16}\text{O}_2$ Laser Transition	$^{12}\text{C}^{16}\text{O}_2$ Laser Transition	(MHz)
$P_{16}(12)$	$R_1(14)$	$R_{11}(34)$	50 566 284.57
$P_{16}(11)$	$R_1(10)$	$R_{11}(14)$	50 676 003.56
$P_{15}(14)$	$R_1(16)$	$R_{11}(12)$	51 088 540.29
$P_{14}(20)$	$R_1(18)$	$R_{11}(16)$	51 132 433.33
$P_{15}(13)$	$R_1(6)$	$P_{11}(18)$	51 201 324.58
$P_{14}(19)$	$R_1(20)$	$R_{11}(16)$	51 252 087.77
$P_{14}(18)$	$R_1(26)$	$R_{11}(30)$	51 370 776.22
$P_{14}(16)$	$R_1(22)$	$R_{11}(6)$	51 605 246.95
$P_{14}(15)$	$R_1(24)$	$R_{11}(6)$	51 721 018.07

a) The frequency values used for ν_1 and ν_2 are taken from Ref. (8) and (12).

b) The estimated uncertainty when used as a transfer oscillator to measure an OCS frequency was less than 0.2 MHz for all entries. The estimated uncertainty (1- σ) of the absolute frequency of each CO line is ± 3 MHz, mainly from errors in determining the CO laser line center. The CO laser was operated at a pressure of 2.67 kPa (20 Torr) with volume percentages 6.3% each for CO, N_2 , and Xe, and balance He.

The uncertainty in both the 26 and 51 THz measurements resulted mainly from the TDL linewidth. TDL linewidths were generally greater in the 51-THz region where widths of 50 to 60 MHz were not uncommon. The numerical value assigned to the uncertainty is essentially one tenth of the beatnote linewidth, plus the Doppler width (HWHM) divided by the signal-to-noise ratio of the derivative signal. A much smaller contribution due to the reference laser frequency uncertainty is also included, but it was never greater than 0.2 MHz.

The measurements were made with an absorption cell having a 1.7-m pathlength, and the OCS gas pressures varied between 27 and 532 Pa (0.2 to 4 Torr). Isotopically enriched samples were used, one contained 90% $^{16}\text{O}^{12}\text{C}^{34}\text{S}$ and the other contained 99% $^{16}\text{O}^{13}\text{C}^{32}\text{S}$. The absorption cell was heated to about 100°C for some of the higher J measurements.

RESULTS AND ANALYSIS

The results of our heterodyne measurements at 26 and 51 THz are summarized in Tables II and III, respectively. These tables also give our estimated experimental uncertainties for each measurement and the differences between the observed frequencies and those calculated from the band constants given later (in Tables V and VI). Two measurements were given an unusually large uncertainty of ± 20 MHz due to the fact that the TDL was not locked to the OCS line. Instead, the measurement was made by sweeping across the absorption line and determining the line center by interpolating between two frequency markers placed on either side of the line center.

TABLE II
Heterodyne Frequency Measurements of Absorption Lines in O¹³CS and
OC³⁴S near 26 THz (860 cm⁻¹)

Molecular Species and Transition	¹³ C ¹⁶ O ₂ Laser Ref. Line ^a	Measured Diff. $\nu_{\text{OCS}} - \nu_{\text{OCO}}$ (MHz)	Measured OCS Frequency ^b (MHz)	Obs.-Calc. (MHz)
¹⁶ O ¹³ C ³² S 20 ⁰ 0 - 10 ⁰ 0	R(15) P(34)	-428.2	25 590 498.9 (10.0)	3.3
	R(25) P(30)	-1095.0	25 703 419.6 (4.0)	1.2
	R(30) P(28)	-1887.2	25 758 468.8 (3.0)	0.6
	R(51) P(20)	1806.1	25 979 208.3 (10.0)	0.2
	R(59) P(17)	-394.4	26 058 761.9 (5.0)	0.2
¹⁶ O ¹² C ³⁴ S 20 ⁰ 0 - 10 ⁰ 0	R(31) P(35)	-804.7	25 572 416.5 (6.0)	2.0
	R(44) P(30)	3044.5	25 707 559.1 (5.0)	1.1
	R(49) P(28)	-2526.0	25 757 830.0 (20.0)	-1.0
	R(55) P(26)	1329.3	25 816 893.7 (5.0)	1.1
¹⁶ O ¹² C ³⁴ S 11 ¹ 0-01 ¹ 0 ^c	R ⁻ (43) P(30)	760.5	25 705 275.1 (4.0)	0.0
	R ⁺ (43) P(30)	2128.5	25 706 643.1 (3.0)	-0.2
	R ⁺ (48) P(28)	-2035.5	25 758 320.5 (20.0)	4.2
	R ⁻ (54) P(26)	1677.4	25 817 241.8 (5.0)	0.0

a) The ¹³CO₂ frequencies in this table are the hot band 01¹1 - [11¹0, 03¹0]₁ lasing transitions (see Ref. (6)).

b) A 2-σ estimated uncertainty in MHz is given in parentheses.

c) R⁻ refers to the e-e transitions and R⁺ refers to the f-f transitions.

TABLE III

Heterodyne Frequency Measurements near 51 THz of the 20⁰0-00⁰ Band Absorption Lines
of Some Isotopes of Carbonyl Sulfide (OCS)

Molecular Species and Transition	¹² C ¹⁶ O Laser Ref. line	Measured Diff. $\nu_{\text{OCS}} - \nu_{\text{CO}}$ (MHz)	Measured OCS Frequency ^a (MHz)	Obs.-Calc. (MHz)
¹⁶ O ¹³ C ³² S	P(34) P ₁₆ (12)	-683.7	50 565 600.9 (6.0)	-1.8
	R(5) P ₁₅ (14)	1082.5	51 089 622.8 (8.0)	-5.2
	R(9) P ₁₄ (20)	3218.9	51 135 652.2 (7.0)	1.3
	R(15) P ₁₅ (13)	1170.4	51 202 495.0 (10.0)	0.3
	R(20) P ₁₄ (19)	4096.3	51 256 184.1 (6.0)	1.4
	R(31) P ₁₄ (18)	-2972.2	51 367 804.0 (4.0)	-3.2
	R(58) P ₁₄ (16)	-1879.4	51 603 367.6 (10.0)	4.9
	R(75) P ₁₄ (15)	1920.4	51 722 938.5 (26.0)	5.1
	¹⁶ O ¹² C ³⁴ S	P(5) P ₁₆ (12)	1329.5	50 567 614.1 (4.0)
R(3) P ₁₆ (11)		-1596.2	50 674 407.4 (6.0)	2.0
R(44) P ₁₅ (14)		-1510.0	51 087 030.3 (7.0)	-3.1
R(58) P ₁₅ (13)		-1638.3	51 199 686.3 (10.0)	-4.2
R(65) P ₁₄ (19)		-1582.8	51 250 505.0 (11.0)	1.0
¹⁶ O ¹² C ³² S		P(52) P ₁₆ (12)	-1761.3	50 564 523.3 (10.0)
	P(16) P ₁₅ (14)	1857.9	51 090 398.2 (7.0)	-3.2
	R(27) P ₁₄ (16)	-784.5	51 604 462.5 (6.0)	5.0
	R(39) P ₁₄ (15)	-1258.3	51 719 759.8 (7.0)	1.1

a) A 2-σ estimated uncertainty in MHz is given in parentheses.

Aside from four measurements of the $11^1_0-01^1_0$ band of the $OC^{34}S$, the present measurements all involved the three energy levels 20^0_0 , 10^0_0 , and 00^0_0 for three isotopic species. Excellent microwave measurements are also available for these levels and the analysis included the appropriate microwave measurements taken from Refs. (9-11). We were also able to make a few less accurate TDL measurements near 26 THz that were calibrated against the $10^0_0-00^0_0$ OCS transitions using the frequencies given in Ref. (4). These more conventional infrared measurements are given in Table IV.

The data for the $l = 0$ states for each isotopic species were combined in a least-squares analysis based on the usual equations

$$\nu_{\text{obs}} = \nu_0 + F'(J) - F''(J''), \quad (2)$$

$$F(J) = B_v J(J+1) - D_v J^2(J+1)^2 + H_v J^3(J+1)^3, \quad (3)$$

where, of course, $\nu_0 = 0$ was appropriate for the microwave transitions, and the H_v term was only used to fit the $^{16}O^{12}C^{32}S$ data.

TABLE IV

Less Accurate Diode Laser Measurements on Carbonyl Sulfide (OCS) near 26 THz. Measurements were Calibrated against $10^0_0-00^0_0$ OCS Frequencies from Ref. (4)

Rotational Transition	$^{16}O^{13}C^{32}S$		Rotational Transition	$^{16}O^{12}C^{34}S$	
	Measured Frequency ^a (MHz)	Obs.-Calc. (MHz)		Measured Frequency ^a (MHz)	Obs.-Calc. (MHz)
$20^0_0-10^0_0$			$20^0_0-10^0_0$		
P(31)	25 010 435(36)	-41	R(12)	25 363 605(52)	33
P(4)	25 353 517(32)	-4	R(19)	25 442 061(50)	-1
P(3)	25 365 737(68)	18	R(20)	25 453 111(32)	-17
R(3)	25 450 083(12)	6	R(21)	25 464 153(20)	-5
R(4)	25 461 988(12)	6	$11^1e_0 - 01^1e_0$		
R(5)	25 473 848(30)	-1	P(7)	25 131 884(35)	-10
R(11)	25 544 251(60)	-30	R(11)	25 355 721(20)	-6
R(12)	25 555 910(32)	19	R(18)	25 435 166(20)	1
R(15)	25 590 500(12)	4	R(19)	25 446 378(72)	-1
R(16)	25 601 958(20)	2	R(22)	25 479 832(30)	15
R(30)	25 758 483(40)	15	R(28)	25 545 735(60)	-44
R(31)	25 769 368(40)	4	$11^1f_0 - 01^1f_0$		
R(32)	25 780 230(36)	8			
R(33)	25 791 046(30)	4	P(7)	25 131 809(35)	-18
R(34)	25 801 824(20)	0	R(11)	25 355 942(20)	1
R(35)	25 812 565(12)	-3	R(12)	25 367 448(60)	35
R(41)	25 876 232(44)	10	R(18)	25 435 573(20)	12
R(51)	25 979 190(50)	-18	R(19)	25 446 804(72)	0
R(52)	25 989 278(20)	-13	R(20)	25 458 010(30)	-4
R(58)	26 048 958(44)	1	R(22)	25 480 338(30)	2
R(59)	26 058 746(56)	-16	R(28)	25 546 484(60)	-22
R(60)	26 068 528(24)	2	R(29)	25 557 418(40)	0
R(61)	26 078 244(12)	-7	R(33)	25 600 738(30)	8
R(79)	26 246 374(60)	46	R(53)	25 809 130(50)	9

a) A 2- σ estimated uncertainty in MHz is given in parentheses after each value.

For completeness, the least-squares analysis included the previously reported TDL measurements of the $10^0\text{--}00^0$ transitions of OC^{34}S and O^{13}CS (5) and the $20^0\text{--}10^0$ and $10^0\text{--}00^0$ transitions of $^{16}\text{O}^{12}\text{C}^{32}\text{S}$ (4). Tables V and VI give the rotational constants and band centers that resulted from this new fit of the data. None of the constants that were reported previously in Refs. (4) and (5) are altered significantly, but the new values and their uncertainties are somewhat more reliable. These new constants are being included in a new calibration paper that will give frequencies, uncertainties, and intensities for the major OCS absorption bands between 800 and 2200 cm^{-1} .

Four of the lines given in Table II are for the $11^1\text{--}01^1\text{--}0$ transitions of OC^{34}S . These measurements were combined with the more conventional measurements calibrated against the $10^0\text{--}00^0$ transitions and given in Table IV. The least-squares analysis of this $l = 1$ band also included microwave measurements taken from Refs. (9) and (10). This analysis used the following equation instead of Eq. (3),

$$F(J) = B_v J(J+1) - D_v [J(J+1) - l^2]^2 \pm 0.5 [q_v J(J+1) - q_{vj} J^2(J+1)^2], \quad (4)$$

where the upper sign for the last term applies to the f levels and the lower sign to the e levels of the l -doublet pair.

AN ESTIMATE OF THE PRESSURE SHIFT

Since other laboratories may use absorption cells with different pathlengths, one can expect that they might also use a pressure of OCS in the absorption cell different from that used in the present measurements. This raises the possibility that pressure-induced shifts may limit the reproducibility of the line frequencies. In order to estimate the magnitude of the pressure shift, we have measured the frequency of the

TABLE V
Rotational Constants for Carbonyl Sulfide (OCS)^a

Molecular Species	Vibrational State	B_v (MHz)	D_v (kHz)	H_v (mHz)	q_v (MHz)	q_{vj} (kHz)
$^{16}\text{O}^{12}\text{C}^{32}\text{S}$	00^0	6081.492117(20) ^b	1.3014398(842)	-0.07197(1368)	-	-
	10^0	6063.358153(387)	1.329507(380)	0.0641(532)	-	-
	20^0	6044.87547(473)	1.359603(1529)	0.1881(1353)	-	-
$^{16}\text{O}^{12}\text{C}^{34}\text{S}$	00^0	5932.833783(164)	1.241274(334)	-	6.06880257(712)	0.0039400(1493)
	01^1	5943.15989(347)	1.24904(1299)	-	-	-
	10^0	5915.14722(175)	1.264697(386)	-	-	-
	11^1	5926.8838(142)	1.28314(1345)	-	6.5078(175)	0.01253(858)
	20^0	5897.12514(535)	1.29018(151)	-	-	-
$^{16}\text{O}^{13}\text{C}^{32}\text{S}$	00^0	6061.923952(240)	1.297827(475)	-	-	-
	10^0	6043.93402(167)	1.327796(507)	-	-	-
	20^0	6025.63276(592)	1.36308(149)	-	-	-

a) To convert to wavenumbers, use the velocity of light: $c = 299792.458\text{ km/s}$.

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

TABLE VI
Band Centers Determined for Carbonyl Sulfide (OCS)

Isotopic Species	Band	Wavenumber (cm^{-1})	Frequency (MHz)
$^{16}\text{C}^{12}\text{C}^{32}\text{S}$	$10^0_0 - 00^0_0$	858.966914(39) ^a	25 751 180.25(118)
	$20^0_0 - 10^0_0$	852.00939(11)	25 542 598.9(34)
	$20^0_0 - 00^0_0$	1710.97630(12)	51 293 779.2(35)
$^{16}\text{O}^{12}\text{C}^{34}\text{S}$	$10^0_0 - 00^0_0$	847.73936(9)	25 414 586.6(27)
	$11^1_0 - 01^1_0$	841.10678(30)	25 215 746.8(90)
	$20^0_0 - 10^0_0$	841.01751(15)	25 213 070.6(45)
	$20^0_0 - 00^0_0$	1688.75687(13)	50 627 657.2(38)
$^{16}\text{O}^{13}\text{C}^{32}\text{S}$	$10^0_0 - 00^0_0$	854.46841(7)	25 616 318.4(21)
	$20^0_0 - 10^0_0$	847.32258(14)	25 402 092.0(43)
	$20^0_0 - 00^0_0$	1701.79099(14)	51 018 410.4(42)

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

$R(29)$ $10^0_0-00^0_0$ transition of $^{16}\text{O}^{12}\text{C}^{32}\text{S}$ at two different pressures. This transition is predicted to lie 2589.9 ± 0.6 MHz higher in frequency than the $P(48)$ laser transition of $^{13}\text{CO}_2$. With OCS at a total pressure of 3 Pa (0.02 Torr) the frequency difference was measured at 2591.0 ± 3.0 MHz. With OCS at a pressure of 1330 Pa (10 Torr) the frequency difference was measured as 2592.2 ± 3.0 MHz. From these measurements we conclude that the pressure shift of the $R(29)$ transition frequency is approximately 0.9 ± 3.8 kHz/Pa (0.1 ± 0.5 MHz/Torr).

There is no reason to expect that the pressure shift should be independent of J (the rotational quantum number), but we would expect that this measurement gives at least the correct order of magnitude for all but the lowest J values for OCS. It would, of course, be useful if more accurate measurements of the pressure shift were made for a wide range of J values.

CONCLUSION

Accurate heterodyne frequency measurements of molecular absorption spectra have been made in the 51-THz region by use of a CO laser calibrated against known CO_2 frequency standards. This opens up the possibility of providing molecular absorption frequency standards far from the CO_2 -laser reference frequencies, but directly connected to the CO_2 frequencies via harmonic generation and mixing measurements with the MIM point-contact diode. Experiments of this type are now underway with a tunable color center laser at 2.3 μm .

Indirect measurements made at 26 THz provided additional information on the same energy levels of OCS and also provided a valuable consistency check on the accuracy of both measurements.

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