The Submillimeter-wave Spectrum of the Formaldehyde Isotopomer $H_2C^{18}O$ in its Ground Vibrational State

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The ground state rotational spectrum of $H_2C^{18}O$ has been studied between 485 and 835 GHz with a sample of natural isotopic composition. Additional lines have been recorded around 130 GHz and near 1.85 THz, using a recently developed far-infrared laser-sideband spectrometer. The accurate new line frequencies were fit together with previously published data to obtain greatly improved spectroscopic constants. Both Watson's S and A reduced Hamiltonians have been employed yielding the rotational constants A_S = 281 961.215 (82), B_S = 36 902.275 51 (36), C_S = 32 513.405 89 (36), A_A = 281 961.371 (82), B_A = 36 904.173 32 (91), and C_A = 32 511.524 65 (86) MHz, respectively.

Key words: Rotational Spectroscopy; Interstellar Molecule; Astrophysics; Astrochemistry; Reduced Hamiltonian.

1. Introduction

Formaldehyde, H_2CO, is one of the most simple and fundamental organic molecules. It is of great atmospheric and astrophysical importance. Its first interstellar detection dates back to 1969 [1], and since then it has even been observed in many external galactic sources. Moreover, several singly substituted isotopomers, such as $H_2{}^{13}CO$ [2], $H_2C^{18}O$ [2], HDCO, and even D_2CO (unlabelled atoms refer to ^{12}C and ^{16}O, respectively) have been observed in the interstellar medium. It has been suggested that $H_2C^{17}O$ and $H_2{}^{13}C^{18}O$ might be observable [3]. Furthermore, some transitions of $H_2{}^{13}CO$ have been identified in the 580 - 720 GHz region in an Orion KL line survey obtained at the Caltech Submillimeter Observatory [4]. This detection suggests that the less abundant $H_2C^{18}O$ should also be observable in that frequency region; overlap by stronger interstellar features might be a problem for the unambiguous identification of this isotopomer. For the extension of radioastronomy to higher frequencies and with the advent of missions such as SOFIA (Stratospheric Observatory For Infrared Astronomy) and FIRST (Far InfraRed Space Telescope), the knowledge of precise rest frequencies is required to unambiguously identify the carriers of spectral features and to investigate the dynamics of the sources *via* Doppler analysis.

Even though the rotational spectrum of formaldehyde has been studied very often, precise laboratory frequencies of several isotopomers up to the low submillimeter region (< 460 GHz) were obtained only quite recently. Since the rotational transitions of formaldehyde isotopomers with an even number of ^1H nuclei follow *a*-type selection rules only, and since distortion effects are fairly large, extrapolation of the rotational spectrum to higher frequencies and higher quantum numbers is risky. Therefore we have started to investigate the ground state rotational spectra of several isotopomers of formaldehyde in the submillimeter and terahertz regions, often supplemented by measurements in the microwave region. Results on H_2CO [5], HDCO [6], and D_2CO [6] up to 2.54, 1.63, and 1.86 THz, respectively, have been reported already. The H_2CO data were further improved by using infrared ground state combination differences in the fit [7]. In the present article, we report accurate transition frequencies of $H_2C^{18}O$ in the submillimeter region, extending previous measurements up to 358 GHz which were summarized by Cornet and Winnewisser [3]. The rotational spectrum of $H_2{}^{13}CO$ has been studied also, and these results will be presented in [8]. Since $H_2C^{18}O$ is a near symmetric prolate top,

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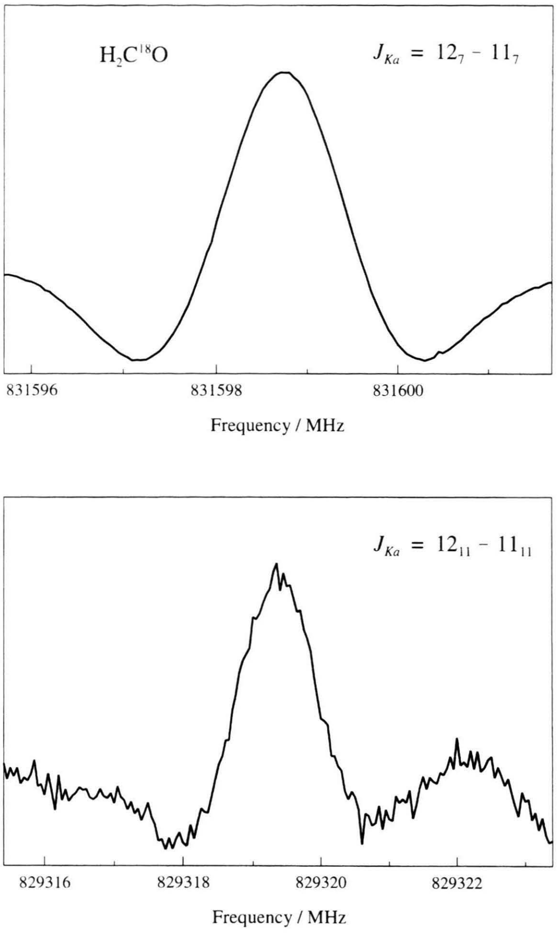

Fig. 1. Two examples of weak submillimeter-wave transitions of $H_2C^{18}O$; the absorption coefficients are $\sim 3.7 \cdot 10^{-4}$ and $\sim 5.3 \cdot 10^{-6}$ cm^{-1}, respectively.

we have tested the performance of Watson's S reduced Hamiltonian in comparison to the A reduced Hamiltonian which was used mostly to fit spectra of the H_2CO isotopomers, and both results are presented.

Table 1. Assignments, frequencies (MHz), uncertainties, and residuals[a] (kHz) of transitions[b] of $H_2C^{18}O$.

$J'_{Ka',Kc'}$-$J''_{Ka'',Kc''}$	Frequency	unc.	o-c(S)	o-c(A)
$3_{3,0}$-$3_{3,1}$	0.4845	1.0	1.0	1.0
$4_{3,1}$-$4_{3,2}$	3.3840	1.0	0.1	0.1
$5_{3,2}$-$5_{3,3}$	13.5310	1.0	–0.1	–0.1
$1_{1,0}$-$1_{1,1}$	4388.79698	0.10	0.02	0.04
$15_{3,12}$-$15_{3,13}$	8744.370	10	–2.9	–3.9
$8_{2,6}$-$8_{2,7}$	12107.035	10	–1.0	3.0
$16_{3,13}$-$16_{3,14}$	12688.997	10	9.7	7.8
$2_{1,1}$-$2_{1,2}$	13165.95491	0.13	–0.02	–0.02
$25_{4,21}$-$25_{4,22}$	13267.205	10	–4.0	–9.5
$35_{5,30}$-$35_{5,31}$	15262.577	10	–0.1	–3.6
$17_{3,14}$-$17_{3,15}$	17971.340	10	–5.3	–8.3
$9_{2,7}$-$9_{2,8}$	18905.493	10	3.2	8.6
$36_{5,31}$-$36_{5,32}$	19905.896	10	3.6	4.4
$27_{4,23}$-$27_{4,24}$	23817.083	10	–6.0	–6.5
$18_{3,15}$-$18_{3,16}$	24901.840	10	5.4	1.1
$37_{5,32}$-$37_{5,33}$	25730.399	10	3.5	7.1
$3_{1,2}$-$3_{1,3}$	26330.140	10	21.4	21.2
$10_{2,8}$-$10_{2,9}$	28120.098	10	–9.3	–2.2
$28_{4,24}$-$28_{4,25}$	31312.809	10	–1.0	4.4
$38_{5,33}$-$38_{5,34}$	32972.319	10	–6.0	–4.9
$19_{3,16}$-$19_{3,17}$	33824.287	10	–0.5	–6.2
$11_{2,9}$-$11_{2,10}$	40179.083	20	–11.7	–2.9
$29_{4,25}$-$29_{4,26}$	40684.809	20	7.0	21.4
$39_{5,34}$-$39_{5,35}$	41898.579	20	6.1	–6.6
$4_{1,3}$-$4_{1,4}$	43878.091	20	44.0	43.3
$20_{3,17}$-$20_{3,18}$	45109.757	20	19.0	12.2
$5_{1,4}$-$5_{1,5}$	65803.150	40	–17.1	–18.7
$1_{0,1}$-$0_{0,0}$	69415.437	10	12.8	12.9

2. Experimental Details

Essentially all accessible transitions between 485 and 835 GHz have been recorded with the Cologne Terahertz Spectrometer with phase-locked BWOs as sources and a magnetically tuned hot electron InSb bolometer as detector. Further details of the spectrometer are given in [9]. Two transitions near 1.87 THz, which were among the strongest of $H_2C^{18}O$, were recorded with a frequency-stabilized FIR laser-sideband spectrometer [10, 11], in which the output of a frequency-locked ring laser (the 1626 GHz laser line of CH_2F_2) is mixed with a phase-locked BWO to generate sideband radiation. For strong single lines, accuracies of better than 20 kHz could be achieved. In the present case, however, the the lines are so weak that the accuracy is limited by the signal-to-noise ratio and by the background features. Finally, two transitions have been recorded near 130 GHz employing a free space absorption cell, an AMC (Analytik Messtechnik Chemnitz) synthesizer as source, and a diode detector. A formaldehyde sample of natural isotopic composition was used throughout.

3. Observed Spectra, Analysis, and Discussion

At lower frequencies (~500 GHz), the new lines of $H_2C^{18}O$ were located close to the predicted positions. The latter were derived from previous data, taking for some higher order centrifugal distortion constants the values of the main isotopomer [7]. The high sensitivity of the spectrometer is demonstrated by the two weak transitions in Figure 1. The non-rigidity of the

Table 1 (continued).

$J'_{Ka',Kc'} - J''_{Ka'',Kc''}$	Frequency	unc.	o-c(S)	o-c(A)
$7_{1,6} - 7_{1,7}$	122730.0447*	12	3.0	–2.8
$2_{1,2} - 1_{1,1}$	134435.9203*	5	2.4	2.4
$2_{0,2} - 1_{0,1}$	138770.861	10	–4.7	–4.6
$2_{1,1} - 1_{1,0}$	143213.068	10	–7.9	–7.9
$3_{0,3} - 2_{0,2}$	208006.441	10	9.6	9.7
$3_{2,2} - 2_{2,1}$	208211.467	10	15.7	15.5
$3_{2,1} - 2_{2,0}$	208444.718	10	–2.4	–2.5
$3_{1,2} - 2_{1,1}$	214778.436	10	9.2	9.1
$4_{0,4} - 3_{0,3}$	277062.652	20	34.6	34.7
$4_{2,3} - 3_{2,2}$	277562.802	10	5.1	4.8
$4_{2,2} - 3_{2,1}$	278145.446	10	2.9	2.8
$4_{1,3} - 3_{1,2}$	286293.689	10	0.6	0.5
$5_{0,5} - 4_{0,4}$	345881.039	20	16.4	16.5
$5_{2,4} - 4_{2,3}$	346869.178	20	–16.8	–17.2
$5_{4} - 4_{4}$	346983.975	20	2.5	1.6
$5_{2,3} - 4_{2,2}$	348032.433	20	25.4	25.5
$5_{1,4} - 4_{1,3}$	357741.049	20	–0.8	–1.1
$7_{2,6} - 6_{2,5}$	485302.411	20	–13.5	–13.8
$7_{6} - 6_{6}$	485380.169	40	–13.2	–12.2
$7_{5} - 6_{5}$	485608.542	20	0.2	–1.0
$7_{4} - 6_{4}$	485843.081	20	–5.8	–7.3
$7_{3,5} - 6_{3,4}$	486125.220	20	19.3	18.4
$7_{3,4} - 6_{3,3}$	486185.962	20	–22.4	–23.2
$7_{1,6} - 6_{1,5}$	500355.758	10	4.7	3.9
$15_{1,14} - 15_{1,15}$	516871.001	80	–65.2	–7.9
$8_{1,8} - 7_{1,7}$	536529.272	15	–6.9	–4.5
$8_{0,8} - 7_{0,7}$	550374.583	10	–15.4	–15.2
$8_{2,7} - 7_{2,6}$	554407.032	15	1.1	0.8
$8_{7} - 7_{7}$	554434.668	20	–9.0	–4.7
$8_{6} - 7_{6}$	554722.572	20	1.0	1.2
$8_{5} - 7_{5}$	554998.795	10	–4.0	–5.6
$8_{3,6} - 7_{3,5}$	555660.932	10	2.2	1.3
$8_{3,5} - 7_{3,4}$	555782.317	15	–9.9	–10.8
$8_{2,6} - 7_{2,5}$	559216.737	20	0.4	1.4
$8_{1,7} - 7_{1,6}$	571481.648	15	–1.0	–2.2
$16_{1,15} - 16_{1,16}$	582847.845	80	–25.1	–63.8
$9_{1,9} - 8_{1,8}$	603231.510	10	–9.4	–6.4
$9_{0,9} - 8_{0,8}$	617740.617	10	4.1	4.3
$9_{4,5} - 8_{4,4}$	624768.928	40	–31.6	–33.5
$9_{3,7} - 8_{3,6}$	625222.998	20	–1.0	–2.0
$9_{2,7} - 8_{2,6}$	630220.745	20	–4.1	–2.8
$9_{1,8} - 8_{1,7}$	642456.260	20	–2.6	–4.2
$10_{1,10} - 9_{1,9}$	669818.260	10	–2.5	1.0
$10_{0,10} - 9_{0,9}$	684662.910	10	–0.7	–0.6
$10_{2,9} - 9_{2,8}$	692337.373	15	–9.5	–9.6
$10_{8} - 9_{8}$	692615.747	30	40.9	47.7
$10_{7} - 9_{7}$	693023.422	10	3.4	5.3
$10_{6} - 9_{6}$	693410.220	20	–2.2	–3.6
$10_{5} - 9_{5}$	693801.223	15	–4.3	–6.8
$10_{4,7} - 9_{4,6}$	694257.504	30	8.2	6.1
$10_{4,6} - 9_{4,5}$	694263.152	30	–6.8	–8.9
$10_{3,8} - 9_{3,7}$	694809.124	15	–1.0	–1.9
$10_{3,7} - 9_{3,6}$	695188.916	10	–4.3	–5.1
$10_{2,8} - 9_{2,7}$	701551.998	10	–2.0	–0.4
$10_{1,9} - 9_{1,8}$	713254.505	10	–7.6	–9.6
$11_{1,11} - 10_{1,10}$	736283.422	15	0.9	4.7
$11_{0,11} - 10_{0,10}$	751137.984	10	3.2	3.1
$11_{10} - 10_{10}$	760829.977	40	6.8	8.2
$11_{2,10} - 10_{2,9}$	761141.648	10	–2.8	–2.8
$11_{9} - 10_{9}$	761361.758	30	7.6	16.2
$11_{8} - 10_{8}$	761853.416	20	–7.1	–1.5
$11_{7} - 10_{7}$	762312.924	20	–6.3	–5.3
$11_{6} - 10_{6}$	762755.688	15	1.0	–0.9
$11_{4,8} - 10_{4,7}$	763770.790	30	31.7	29.8
$11_{4,7} - 10_{4,6}$	763782.036	30	–32.5	–34.4
$11_{3,9} - 10_{3,8}$	764415.312	10	3.3	2.6
$11_{3,8} - 10_{3,7}$	765030.350	10	8.1	7.5
$11_{2,9} - 10_{2,8}$	773200.623	30	–15.2	–13.5
$11_{1,10} - 10_{1,9}$	783849.217	10	–4.5	–6.8
$19_{1,18} - 19_{1,19}$	797402.966	60	18.0	47.0
$12_{1,12} - 11_{1,11}$	802622.681	10	–2.1	1.6
$12_{0,12} - 11_{0,11}$	817180.342	15	8.3	7.7
$12_{11} - 11_{11}$	829319.369	50	3.3	–29.8
$12_{2,11} - 11_{2,10}$	829824.722	15	8.9	9.1
$12_{10} - 11_{10}$	829952.697	80	52.9	55.2
$12_{9} - 11_{9}$	830538.952	80	–79.6	–70.9
$12_{8} - 11_{8}$	831084.264	40	–29.9	–24.6
$12_{7} - 11_{7}$	831598.731	10	–6.5	–5.6
$12_{6} - 11_{6}$	832102.377	15	3.0	1.3
$12_{5} - 11_{5}$	832638.635	10	6.2	3.9
$12_{4,9} - 11_{4,8}$	833307.721	15	2.7	1.1
$12_{4,8} - 11_{4,7}$	833328.892	15	4.0	2.4
$12_{3,10} - 11_{3,9}$	834035.807	10	6.7	6.3
$12_{3,9} - 11_{3,8}$	834988.240	10	4.1	3.7
$27_{1,26} - 26_{1,25}$	1862487.065	200	31.9	–20.6
$27_{3,25} - 26_{3,24}$	1872169.057	200	–93.6	–37.3

[a] o-c(S) and o-c(A) refer to the residuals obtained with the S and A reduced Hamiltonian, respectively. [b] Frequencies up to 358 GHz, except two lines marked by asterisks, from Cornet and Winnewisser [3], all others from this work. Since $K_a + K_c = J$ or $J + 1$, K_c has been omitted for prolate paired transitions.

molecule was apparent from the fact that lines were often lying well outside the sum of the experimental and predicted uncertainties, sometimes even at low K_a. Despite the rather sparse spectrum, some lines of $H_2C^{18}O$ were overlapped by other lines, e. g. of other isotopomers. These lines were recognized by a much larger intensity than expected or by differences between experimental and calculated positions that were much larger than the estimated uncertainty; they were not included in the final line-list given in Table 1. One example for an overlapped line was found at 1 848 551.902 (200) MHz, one third of which turned out to be due to $28_{1,28}$ - $27_{1,27}$ of $H_2C^{18}O$ and about two thirds to $26_{5,22}$ - $25_{5,21}$ of $H_2{}^{13}CO$.

Formaldehyde is a near symmetric, prolate rotor with an asymmetry parameter $\kappa = -0.9648$ for $H_2C^{18}O$. Therefore, Watson's S reduction of the rotational Hamiltonian should be more appropriate than

Table 2. Rotational and centrifugal distortion constants[a] of $H_2C^{18}O$ in Watson's S reduction compared to values of H_2CO [7], standard deviation rms (MHz) and dimensionless weighted rms.

	$H_2C^{18}O$	H_2CO
A	281 961 .215 (82)	281 970 .540 2 (101)
B	36 902 .275 51 (36)	38 833 .987 49 (47)
C	32 513 .405 89 (36)	34 004 .244 07 (45)
$D_J \cdot 10^3$	64 .306 34 (218)	70 .324 96 (180)
$D_{JK} \cdot 10^3$	1 202 .078 (25)	1 321 .001 (42)
D_K	19 .459 (39)	19 .390 50 (99)
$d_1 \cdot 10^3$	−9 .082 19 (62)	−10 .437 621 (260)
$d_2 \cdot 10^3$	−2 .076 42 (96)	−2 .501 385 (81)
$H_J \cdot 10^9$	6 .64 (255)	3 .20 (165)
$H_{JK} \cdot 10^6$	6 .080 (98)	7 .105 (72)
$H_{KJ} \cdot 10^6$	1 .703 (186)	9 .46 (117)
$H_K \cdot 10^3$	4 .018 5	4 .018 5 (280)
$h_1 \cdot 10^9$	27 .59 (164)	31 .830 (256)
$h_2 \cdot 10^9$	37 .09 (95)	47 .350 (223)
$h_3 \cdot 10^9$	12 .640 (131)	15 .970 (40)
$L_{JK} \cdot 10^9$	6 .59	6 .59 (218)
$L_{KKJ} \cdot 10^9$	−10 .7	−10 .7 (46)
$L_K \cdot 10^9$	−588	−588 (202)
$l_3 \cdot 10^{15}$	−374	−374 (51)
$l_4 \cdot 10^{15}$	−147 .0	−147 .0 (98)
rms $\cdot 10^3$	18 .64	
wrms	0 .664	

[a] Numbers in parentheses are one standard deviation in units of the least significant figures.

Table 3. Rotational and centrifugal distortion constants[a] of $H_2C^{18}O$ in Watson's A reduction compared to values of H_2CO [7], standard deviation rms (MHz) and dimensionless weighted rms.

	$H_2C^{18}O$	H_2CO
A	281 961.371 (82)	281 970.522 1 (103)
B	36 904.173 32 (91)	38 836.050 64 (48)
C	32 511.524 65 (86)	34 002.200 41 (46)
$\Delta_J \cdot 10^3$	68.460 31 (226)	75.325 17 (179)
$\Delta_{JK} \cdot 10^3$	1 177.244 (24)	1 291.119 (40)
Δ_K	19.524 (39)	19.415 09 (99)
$\delta_J \cdot 10^3$	9.095 323 (283)	10.454 476 (234)
$\delta_K \cdot 10^3$	946.20 (40)	1 028.115 (56)
$\Phi_J \cdot 10^9$	85.0 (34)	97.93 (172)
$\Phi_{JK} \cdot 10^6$	27.984 (257)	32.398 (111)
$\Phi_{KJ} \cdot 10^6$	−73.02 (81)	−76.69 (109)
$\Phi_K \cdot 10^3$	4.074 9	4.074 9 (280)
$\phi_J \cdot 10^9$	36.64 (171)	44.535 (283)
$\phi_{JK} \cdot 10^6$	14.37 (43)	17.162 (66)
$\phi_K \cdot 10^6$	1 427.2 (181)	1 491.0 (40)
$L_{JJK} \cdot 10^9$	−1.595	−1.595 (62)
$L_{JK} \cdot 10^9$	−5.97	−5.97 (204)
$L_{KKJ} \cdot 10^9$	41	41.0 (46)
$L_K \cdot 10^9$	−607	−607(202)
$l_{KJ} \cdot 10^9$	−85.5	−85.5 (34)
rms	19.86	
wrms	0.692	

[a] Numbers in parentheses are one standard deviation in units of the least significant figures.

the more widely used A reduction. Infact, in the course of the investigation it was found that often predictions and fits in the S reduction were better than those in the A reduction, as judged by the standard deviations obtained in the fitting process. However, as for the main isotopomer [7], the differences in the quality of the fit between the S and the A reductions were generally small, and occasionally smaller residuals were obtained with the A reduction. Similar to the main isotopomer, the coefficient s_{111} of the unitary transformation for the A reduction, $\sim 6 \cdot 10^{-7}$, is smaller than $D/B \approx 1.86 \cdot 10^{-6}$, ensuring fast convergence of the rotational Hamiltonian for moderate values of J and K_a.

It is well known that for a large number of experimental lines of a fairly floppy molecule there does not exist a unique set of spectroscopic constants to be used in the fit. In our case, constants that significantly reduced the standard deviation and that were well determined, i. e. with a value at least three times larger than the uncertainty, were retained in the fit. As can be seen in Table 1, some higher order constants were kept fixed to values of the main isotopomer [7]. For the S fit, H_J was fitted also because its value and its uncertainty are reasonable and because its uncertainty for the main isotopomer is not much smaller.

The obtained rotational and centrifugal distortion constants of $H_2C^{18}O$ are presented in Tables 2 and 3 together with values for the main isotopomer. The complete fit file, including correlation coefficients, as well as new line predictions for astrophysical purpose are available online from the Cologne Database for Molecular Spectroscopy (http://www.ph1.uni-koeln.de/vorhersagen/).

In general, the spectroscopic constants of $H_2C^{18}O$ are smaller than those of H_2CO because of the larger mass of ^{18}O compared to ^{16}O; the constants D_K and Φ_K, respectively, are a possible exception. In most cases, the changes between the two isotopomers are comparable to the changes in appropriate powers of the rotational constants. Some larger differences, such as for H_{KJ} and h_2, may be explained by the small value of the constants compared to related ones. The smaller uncertainties of some $H_2C^{18}O$ constants compared to those of

H_2CO are most likely caused by the smaller number of distortion constants determined. As for the main isotopomer, Watson's S reduction results in a final fit just slightly better than the A reduction. Thus, the use of the A reduction for fitting and predicting spectra of isotopomers of formaldehyde is about as reasonable as using the S reduction. Table 1 shows also that the residuals obtained with the A and the S reduced Hamiltonians are in general very similar; occasionally, however, larger deviations are observed, but they do not seem to be systematic.

With the present investigation, the data set of $H_2C^{18}O$ has been enlarged substantially. As a consequence, improved spectroscopic constants have been obtained which allow the strongest transitions (^qR-branch transitions with $K_a \lesssim 3$) to be predicted which uncertainties of less than 1 MHz up to ~2 THz.

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