JOURNAL OF MOLECULAR SPECTROSCOPY 171, 481–493 (1995)

Millimeter- and Submillimeter-Wave Rotational Spectra of Rare Hydrogen Sulfide Isotopomers

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Pure rotational transitions of the hydrogen sulfide isotopomers $H_2{}^{33}S$, $H_2{}^{34}S$, and $H_2{}^{36}S$ have been recorded with high resolution in the frequency range between 165 and 1072 GHz in natural abundance. Together with previously reported FIR pure rotational transitions of $H_2{}^{33}S$ and $H_2{}^{34}S$, these data are used to derive an improved set of rotational and centrifugal distortion constants. For the $H_2{}^{33}S$ species, electric quadrupole and magnetic spin–rotation hfs constants are also determined. The new data allow accurate frequency predictions throughout the submillimeter-wave region for the two isotopomers $H_2{}^{34}S$ and $H_2{}^{33}S$, including hyperfine splittings for $H_2{}^{33}S$. These predictions are of astrophysical relevance.

I. INTRODUCTION

High-resolution scanning spectroscopy in the terahertz region has recently been accomplished in our Cologne laboratory by frequency- and phase-stabilization of continuously tunable high frequency backward wave oscillators (BWOs) (*1*). The present paper is part of an effort to measure the rotational spectra of astrophysically important molecules into the terahertz region, with the aim of providing highly accurate frequency predictions usable for astrophysical identifications required in line surveys or dedicated molecular line searches. In the present paper we report rotational spectra of the ^{33}S, ^{34}S, and ^{36}S isotopomers of H_2S.

In 1953, Burrus and Gordy observed the first transitions in the millimeter-wave (mm-wave) region of H_2S, including the sulfur isotopomers $H_2{}^{34}S$ and $H_2{}^{33}S$ (*2*). About 20 years later, a thorough study of H_2S and its deuterated species up to 760 GHz was published by Helminger *et al.* (*3*). For $H_2{}^{34}S$ and $H_2{}^{33}S$ only the $1_{1,0}$–$1_{0,1}$ and $2_{2,0}$–$2_{1,1}$ transitions have been reported in the literature (*2–5*). To our knowledge, no data on the $H_2{}^{36}S$ isotopomer with 0.02% relative abundance have been reported thus far. FIR pure rotational transitions of H_2S, $H_2{}^{34}S$, and $H_2{}^{33}S$ have been observed between 1.8 and 9 THz by Flaud *et al.* (*6*), using Fourier transform techniques. These measurements supplied the basis for the first frequency predictions of the ^{33}S- and ^{34}S-substituted species in the mm- and submillimeter-wave (submm-wave) region.

In the interstellar medium (ISM), H_2S was first detected by Thaddeus *et al.* (*7*) in seven galactic sources. Minh *et al.* (*8*) observed H_2S in the cold, dark clouds L134N and TMC 1. Furthermore, they detected H_2S and $H_2{}^{34}S$ toward several positions in OMC 1 (*9*) as well as toward several star-forming regions (*10*), where high excitation

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0022-2852/95 $6.00

TABLE I

$H_2{}^{34}S$: Observed Rotational Transitions, Together with the Predicted Spectrum ($J \leqslant 10$) up to 1100 GHz (Calculated from the Constants of Table IV)

J'	K'_a	K'_c	←	J''	K''_a	K''_c	ν_{exp}	$\Delta\nu_{exp}$ [a]	ν_{calc}	σ [b]	E''
3	3	1		4	0	4			30 895.72	0.06	113.990
7	1	6		6	4	3	89 756.561	0.050	89 756.59	0.03	358.780
4	2	2		5	1	5			122 492.60	0.11	166.077
10	2	8		9	5	5			129 384.34	0.88	737.359
10	3	8		9	4	5			145 932.60	0.80	736.807
7	3	4		8	2	7	163 860.763	0.050	163 860.78	0.04	451.771
1	1	0		1	0	1	167 910.516 [c]	0.002	167 910.52	0.00	13.739
7	2	6		6	3	3	170 149.398	0.050	170 149.40	0.03	356.099
7	4	4		8	1	7			185 662.29	0.09	451.771
4	1	4		3	2	1			201 889.20	0.05	107.260
2	2	0		2	1	1	214 376.924 [c]	0.002	214 376.92	0.00	51.096
4	3	2		5	0	5			226 700.09	0.12	166.076
3	3	0		3	2	1	295 431.052	0.090	295 431.06	0.01	107.260
6	1	5		5	4	2			317 145.61	0.08	270.569
4	3	1		4	2	2			367 830.67	0.04	170.163
3	2	1		3	1	2	369 246.090	0.160	369 246.11	0.02	94.943
3	1	3		2	2	0			392 811.46	0.03	58.247
2	1	1		2	0	2	393 725.700	0.160	393 725.73	0.01	37.963
5	4	1		5	3	2			403 338.97	0.05	263.492
9	2	7		8	5	4			408 781.36	0.54	609.791
4	4	0		4	3	1	415 359.190	0.100	415 359.11	0.03	182.433
5	1	4		4	4	1			443 146.33	0.07	195.106
1	1	1		0	0	0	451 081.597	0.100	451 081.61	0.02	0.000
5	2	3		6	1	6			462 825.87	0.22	227.579
9	3	7		8	4	4			468 699.37	0.47	607.793
8	3	5		9	2	8			471 105.92	0.17	551.130
8	4	5		9	1	8			476 136.41	0.23	551.130
6	5	1		6	4	2			484 372.77	0.05	375.073
5	3	3		6	0	6			492 270.66	0.22	227.579
2	2	1		2	1	2	503 013.679	0.100	503 013.65	0.01	38.231
6	2	5		5	3	2			529 504.24	0.06	263.492
7	5	2		7	4	3	554 043.448	0.100	554 043.51	0.05	486.741
3	3	1		3	2	2	563 676.279	0.100	563 676.39	0.02	96.218
5	5	0		5	4	1	566 665.836	0.100	566 665.83	0.05	276.946
6	4	2		6	3	3	568 816.665	0.100	568 816.70	0.03	356.099
8	6	2		8	5	3			586 708.11	0.08	634.818
7	6	1		7	5	2	611 883.600	0.100	611 883.61	0.06	505.222
5	3	2		5	2	3	613 834.575	0.100	613 834.57	0.03	243.017
4	4	1		4	3	2	643 590.281	0.100	643 590.29	0.04	173.638
8	2	6		7	5	3			657 675.45	0.27	492.541
4	2	2		4	1	3	666 816.195	0.100	666 816.26	0.03	147.920
9	7	2		9	6	3	675 635.018	0.100	675 634.99	0.08	800.538
2	0	2		1	1	1	687 024.663	0.200	687 024.79	0.03	15.046
3	1	2		3	0	3			708 457.31	0.02	71.312
10	7	3		10	6	4			731 287.86	1.21	959.956
6	6	0		6	5	1			731 595.40	0.07	391.230
2	1	2		1	0	1	734 268.688	0.100	734 268.72	0.03	13.739
5	5	1		5	4	2			739 796.55	0.05	270.569
3	2	2		3	1	3			745 519.54	0.02	71.350
9	6	3		9	5	4			750 262.36	0.44	775.512
4	3	2		4	2	3			763 228.30	0.04	148.180
9	3	6		10	2	9			767 320.93	0.58	659.838
9	4	6		10	1	9			768 417.69	0.63	659.838
8	7	1		8	6	2			774 485.65	0.11	654.388
6	2	4		7	1	7			777 685.03	0.41	298.489
6	3	4		7	0	7			783 982.56	0.41	298.489
5	4	2		5	3	3			796 541.75	0.05	243.999
8	5	3		8	4	4			810 163.44	0.19	607.793
10	8	2		10	7	3			818 277.06	0.69	984.350
5	2	4		4	3	1			824 401.44	0.07	182.433
8	3	6		7	4	3			831 785.12	0.27	486.741
7	2	5		6	5	2			832 061.85	0.14	387.078
6	6	1		6	5	2			847 419.76	0.07	387.078
6	5	2		6	4	3			848 354.94	0.05	358.780
6	2	4		5	5	1			874 914.41	0.09	295.246
7	4	3		7	3	4			884 523.67	0.09	457.237
7	7	0		7	6	1			891 479.86	0.09	525.633

Note. Frequencies given in MHz. E'' given in cm^{-1}.

[a] Estimated measurement uncertainty.

[b] One standard deviation of the calculated frequency.

[c] From Ref. (5).

TABLE I—*Continued*

J'	K'_a	K'_c	←	J''	K''_a	K''_c	ν_{exp}	$\Delta\nu_{exp}$	ν_{calc}	σ	E''
7	6	2		7	5	3			919 142.03	0.06	492.541
4	2	3		3	3	0	931 307.140	0.100	931 307.02	0.05	117.115
6	3	3		6	2	4	949 424.889	0.100	949 424.76	0.04	324.430
9	8	1		9	7	2	950 623.090	0.100	950 623.12	0.12	823.075
7	7	1		7	6	2	960 381.869	0.100	960 381.86	0.08	523.201
3	0	3		2	1	2	991 732.888	0.100	991 732.90	0.04	38.231
10	3	7		9	6	4			993 063.59	1.28	780.065
5	2	3		5	1	4	993 180.302	0.100	993 180.37	0.03	209.888
3	1	3		2	0	2	1 000 914.138	0.100	1 000 914.12	0.03	37.963
8	7	2		8	6	3	1 006 715.596	0.100	1 006 715.55	0.07	645.334
4	1	3		4	0	4	1 017 221.754	0.100	1 017 221.75	0.05	113.990
5	3	3		5	2	4	1 021 314.917	0.100	1 021 314.97	0.04	209.932
6	4	3		6	3	4	1 023 486.614	0.100	1 023 486.59	0.04	324.640
4	2	3		4	1	4	1 024 848.913	0.100	1 024 848.88	0.05	113.995
8	8	0		8	7	1	1 035 127.066	0.100	1 035 127.07	0.11	680.222
7	5	3		7	4	4	1 036 608.627	0.100	1 036 608.58	0.04	457.964
8	6	3		8	5	4	1 065 572.053	0.100	1 065 572.13	0.07	609.791
2	2	1		1	1	0	1 069 371.817	0.100	1 069 371.85	0.03	19.340
10	6	4		10	5	5	1 071 578.823	0.100	1 071 578.83	0.08	924.212
8	8	1		8	7	2	1 072 501.022	0.100	1 072 501.03	0.10	678.915
7	2	5		8	1	8			1 080 304.37	0.74	378.797
7	3	5		8	0	8			1 081 519.21	0.73	378.797

conditions prevail. All these interstellar observations were carried out by using the $1_{1,0}-1_{0,1}$ transition of the main isotope and the $H_2{}^{34}S$ isotopomer. However, up to now, $H_2{}^{33}S$ has not been detected in the ISM. Yet the constantly increasing sensitivity of the receiving equipment, both in mixer-elements and telescopes, has resulted in the detection of less abundant molecular species, as summarized by Winnewisser *et al.* (*11*). Replacement of the Schottky diode mixer (*12*) with superconductor–insulator–superconductor (SIS) tunnel junctions, but with a present frequency limit at about 700 GHz, as the most critical item in any receiver (*13*), warrants laboratory as well as astrophysical investigation of molecular spectra up to the terahertz region. Especially the unambiguous assignment of unidentified interstellar lines makes knowledge of highly precise laboratory rest frequencies imperative. Hereby rotational spectra of vibrationally excited states of known interstellar molecules are equally relevant as spectra of the less abundant isotopomers. Thus, accurate rest frequencies are also highly desirable for the $H_2{}^{34}S$ and $H_2{}^{33}S$ species.

In this paper, we present high-resolution mm- and submm-wave measurements of $H_2{}^{34}S$, $H_2{}^{33}S$, and $H_2{}^{36}S$ in the frequency range between 165 and 1072 GHz. In the analysis, rotational constants are derived for $H_2{}^{34}S$ and $H_2{}^{33}S$ as well as quartic, sextic, and octic centrifugal constants by combining our data with the FIR data from Ref. (*6*). For $H_2{}^{33}S$, hyperfine parameters of the electric quadrupole and the magnetic spin–rotation interaction are also given. The new sets of molecular constants provide firm frequency predictions for $H_2{}^{34}S$ and $H_2{}^{33}S$ in the mm- and submm-wave region, including, for the first time, the hyperfine splittings due to the ^{33}S nucleus.

II. EXPERIMENTAL DETAILS

The mm- and submm-wave spectrometer at Cologne University was used in two different configurations. Up to 370 GHz, a multiplied Gunn oscillator was used as the frequency source. Between 370 and 1072 GHz, the frequencies were generated by

TABLE II

$H_2{}^{33}S$: Observed Rotational Transitions, Together with the Predicted Spectrum ($J \leq 10$) up to 1100 GHz (Calculated from the Constants of Tables IV and V)

J'	K'_a	K'_c	F'	←	J''	K''_a	K''_c	F''	ν_{exp}	$\Delta\nu_{exp}$[a]	ν_{calc}	σ[b]	E''	rel. Int.
3	3	1	1.5		4	0	4	2.5			32 874.00	0.09	114.078	0.143
3	3	1	4.5		4	0	4	5.5			32 884.09	0.06	114.078	0.333
3	3	1	2.5		4	0	4	3.5			32 899.52	0.07	114.078	0.191
3	3	1	3.5		4	0	4	4.5			32 909.71	0.06	114.078	0.255
7	1	6	7.5		6	4	3	6.5			89 628.61	0.57	359.064	0.259
7	1	6	6.5		6	4	3	5.5			89 631.38	0.57	359.064	0.223
7	1	6	8.5		6	4	3	7.5			89 641.67	0.57	359.064	0.300
7	1	6	5.5		6	4	3	4.5			89 644.44	0.58	359.064	0.192
4	2	2	2.5		5	1	5	3.5			121 114.85	0.21	166.207	0.167
4	2	2	5.5		5	1	5	6.5			121 120.19	0.17	166.207	0.318
4	2	2	3.5		5	1	5	4.5			121 132.85	0.19	166.206	0.207
4	2	2	4.5		5	1	5	5.5			121 138.11	0.18	166.206	0.258
10	2	8	10.5		9	5	5	9.5			130 371.16	3.35	737.896	0.258
10	2	8	9.5		9	5	5	8.5			130 372.51	3.35	737.896	0.233
10	2	8	11.5		9	5	5	10.5			130 380.24	3.35	737.896	0.286
10	2	8	8.5		9	5	5	7.5			130 381.60	3.35	737.896	0.211
10	3	8	10.5		9	4	5	9.5			147 705.23	3.12	737.318	0.258
10	3	8	9.5		9	4	5	8.5			147 706.56	3.12	737.318	0.233
10	3	8	11.5		9	4	5	10.5			147 714.20	3.12	737.318	0.286
10	3	8	8.5		9	4	5	7.5			147 715.58	3.12	737.318	0.211
7	3	4	5.5		8	2	7	6.5			162 696.85	1.01	452.122	0.200
7	3	4	8.5		8	2	7	9.5			162 699.03	1.01	452.122	0.294
7	3	4	6.5		8	2	7	7.5			162 708.57	1.01	452.121	0.228
7	3	4	7.5		8	2	7	8.5			162 710.71	1.01	452.121	0.259
1	1	0	0.5		1	0	1	0.5	168 304.408	0.060	168 304.43	0.03	13.743	0.028
1	1	0	0.5		1	0	1	1.5	168 319.216	0.160	168 319.14	0.02	13.742	0.139
1	1	0	2.5		1	0	1	2.5	168 319.216	0.160	168 319.25	0.02	13.742	0.350
1	1	0	1.5		1	0	1	0.5	168 323.115	0.060	168 323.11	0.02	13.743	0.139
1	1	0	2.5		1	0	1	1.5	168 327.561	0.060	168 327.55	0.02	13.742	0.150
1	1	0	1.5		1	0	1	2.5	168 329.515	0.060	168 329.53	0.02	13.742	0.150
1	1	0	1.5		1	0	1	1.5	168 337.823	0.060	168 337.83	0.02	13.742	0.044
7	2	6	7.5		6	3	3	6.5			172 479.87	0.59	356.301	0.259
7	2	6	6.5		6	3	3	5.5			172 482.48	0.59	356.301	0.223
7	2	6	8.5		6	3	3	7.5			172 492.08	0.59	356.301	0.300
7	2	6	5.5		6	3	3	4.5			172 494.76	0.60	356.301	0.192
7	4	4	5.5		8	1	7	6.5			185 385.69	1.00	452.122	0.200
7	4	4	8.5		8	1	7	9.5			185 387.87	1.00	452.122	0.294
7	4	4	6.5		8	1	7	7.5			185 397.58	1.00	452.121	0.228
7	4	4	7.5		8	1	7	8.5			185 399.75	0.99	452.121	0.259
4	1	4	4.5		3	2	1	3.5			202 969.92	0.06	107.313	0.255
4	1	4	3.5		3	2	1	2.5			202 977.37	0.07	107.313	0.191
4	1	4	5.5		3	2	1	4.5			202 989.42	0.07	107.312	0.333
4	1	4	2.5		3	2	1	1.5			202 996.93	0.09	107.312	0.143
2	2	0	0.5		2	1	1	1.5	215 494.413	0.090	215 494.45	0.02	51.118	0.050
2	2	0	0.5		2	1	1	0.5	215 496.651	0.090	215 496.66	0.02	51.118	0.050
2	2	0	3.5		2	1	1	2.5	215 500.805	0.090	215 500.79	0.03	51.118	0.057
2	2	0	3.5		2	1	1	3.5	215 502.804	0.090	215 502.80	0.02	51.118	0.343
2	2	0	1.5		2	1	1	2.5	215 503.820	0.090	215 503.74	0.02	51.118	0.070
2	2	0	1.5		2	1	1	1.5	215 505.387	0.090	215 505.37	0.01	51.118	0.080
2	2	0	1.5		2	1	1	0.5	215 507.589	0.090	215 507.58	0.02	51.118	0.050
2	2	0	2.5		2	1	1	2.5	215 511.584	0.090	215 511.57	0.02	51.118	0.173
2	2	0	2.5		2	1	1	1.5	215 513.395	0.400	215 513.21	0.02	51.118	0.070
2	2	0	2.5		2	1	1	3.5	215 513.395	0.400	215 513.58	0.03	51.118	0.057
4	3	2	2.5		5	0	5	3.5			227 580.95	0.20	166.206	0.167
4	3	2	5.5		5	0	5	6.5			227 586.83	0.16	166.206	0.318
4	3	2	3.5		5	0	5	4.5			227 600.84	0.18	166.206	0.207
4	3	2	4.5		5	0	5	5.5			227 606.73	0.17	166.206	0.258
3	3	0	1.5		3	2	1	2.5	297 874.039	0.120	297 874.03	0.03	107.313	0.029
3	3	0	4.5		3	2	1	3.5	297 877.216	0.120	297 877.24	0.03	107.313	0.030
3	3	0	1.5		3	2	1	1.5	297 879.065	0.120	297 879.05	0.02	107.312	0.114
3	3	0	4.5		3	2	1	4.5	297 881.995	0.120	297 881.99	0.03	107.312	0.327
3	3	0	2.5		3	2	1	3.5	297 883.320	0.120	297 883.22	0.03	107.313	0.038
3	3	0	2.5		3	2	1	2.5	297 885.623	0.120	297 885.66	0.02	107.313	0.147
3	3	0	3.5		3	2	1	3.5	297 888.680	0.120	297 888.71	0.02	107.313	0.218
3	3	0	2.5		3	2	1	1.5	297 890.480	0.120	297 890.69	0.03	107.312	0.029
3	3	0	3.5		3	2	1	2.5	297 890.979	0.120	297 891.14	0.03	107.313	0.038
3	3	0	3.5		3	2	1	4.5	297 893.332	0.120	297 893.46	0.03	107.312	0.030
6	1	5	6.5		5	4	2	5.5			315 835.49	0.30	270.829	0.259
6	1	5	5.5		5	4	2	4.5			315 839.29	0.30	270.829	0.217
6	1	5	7.5		5	4	2	6.5			315 850.45	0.30	270.829	0.308
6	1	5	4.5		5	4	2	3.5			315 854.20	0.31	270.829	0.182
4	3	1	2.5		4	2	2	2.5	368 443.148	0.160	368 443.26	0.04	170.247	0.149
4	3	1	5.5		4	2	2	5.5	368 444.700	0.160	368 444.64	0.04	170.247	0.315
4	3	1	3.5		4	2	2	3.5	368 447.240	0.160	368 447.21	0.04	170.247	0.181
4	3	1	4.5		4	2	2	4.5	368 448.664	0.160	368 448.59	0.04	170.247	0.236
3	2	1	1.5		3	1	2	1.5	369 165.613	0.160	369 165.39	0.03	94.998	0.114
3	2	1	4.5		3	1	2	4.5	369 168.170	0.160	369 168.31	0.03	94.998	0.327
3	2	1	2.5		3	1	2	2.5	369 171.560	0.160	369 171.55	0.03	94.998	0.147
3	2	1	3.5		3	1	2	3.5	369 174.315	0.160	369 174.43	0.03	94.998	0.218

Note. Frequencies given in MHz. E'' in cm^{-1}. Hyperfine components with relative intensities below 0.05 have been omitted.

[a] Estimated measurement uncertainty.

[b] One standard deviation of the calculated frequency.

TABLE II—*Continued*

J'	K'_a	K'_c	F'	←	J''	K''_a	K''_c	F''	ν_{exp}	$\Delta\nu_{exp}$	ν_{calc}	σ	E''	rel. Int.
3	1	3	3.5		2	2	0	2.5			392 703.94	0.05	58.306	0.245
3	1	3	2.5		2	2	0	2.5			392 709.96	0.05	58.306	0.052
3	1	3	2.5		2	2	0	1.5			392 717.80	0.05	58.306	0.160
3	1	3	4.5		2	2	0	3.5			392 727.92	0.06	58.306	0.357
3	1	3	1.5		2	2	0	0.5			392 741.74	0.07	58.306	0.100
2	1	1	0.5		2	0	2	0.5			393 577.93	0.06	37.989	0.050
2	1	1	1.5		2	0	2	0.5			393 580.13	0.05	37.989	0.050
2	1	1	3.5		2	0	2	3.5			393 587.32	0.05	37.989	0.343
2	1	1	0.5		2	0	2	1.5			393 588.76	0.05	37.989	0.050
2	1	1	2.5		2	0	2	3.5			393 589.33	0.05	37.989	0.057
2	1	1	1.5		2	0	2	1.5			393 590.97	0.04	37.989	0.080
2	1	1	2.5		2	0	2	1.5			393 592.60	0.05	37.989	0.070
2	1	1	3.5		2	0	2	2.5			393 598.29	0.05	37.989	0.057
2	1	1	1.5		2	0	2	2.5			393 598.67	0.05	37.989	0.070
2	1	1	2.5		2	0	2	2.5			393 600.30	0.05	37.989	0.173
5	4	1	3.5		5	3	2	3.5			405 417.14	0.06	263.612	0.170
5	4	1	6.5		5	3	2	6.5			405 417.98	0.06	263.612	0.306
5	4	1	4.5		5	3	2	4.5			405 420.37	0.06	263.612	0.199
5	4	1	5.5		5	3	2	5.5			405 421.27	0.06	263.612	0.244
9	2	7	9.5		8	5	4	8.5			409 548.15	1.88	610.243	0.259
9	2	7	8.5		8	5	4	7.5			409 549.75	1.88	610.243	0.231
9	2	7	10.5		8	5	4	9.5			409 557.80	1.88	610.243	0.289
9	2	7	7.5		8	5	4	6.5			409 559.42	1.88	610.243	0.206
4	4	0	2.5		4	3	1	3.5	419 681.135	0.250	419 681.05	0.03	182.537	0.018
4	4	0	5.5		4	3	1	4.5	419 683.431	0.250	419 683.25	0.04	182.537	0.018
4	4	0	2.5		4	3	1	2.5	419 687.459	0.100	419 687.47	0.03	182.537	0.149
4	4	0	5.5		4	3	1	5.5	419 689.418	0.100	419 689.33	0.03	182.537	0.315
4	4	0	3.5		4	3	1	3.5	419 693.434	0.100	419 693.40	0.02	182.537	0.181
4	4	0	4.5		4	3	1	4.5	419 695.479	0.100	419 695.43	0.02	182.537	0.236
4	4	0	4.5		4	3	1	3.5	419 697.878	0.150	419 697.80	0.03	182.537	0.024
4	4	0	3.5		4	3	1	2.5	419 699.892	0.150	419 699.82	0.03	182.537	0.018
4	4	0	4.5		4	3	1	5.5	419 701.559	0.150	419 701.51	0.04	182.537	0.018
5	1	4	5.5		4	4	1	4.5			439 863.48	0.12	195.375	0.258
5	1	4	4.5		4	4	1	3.5			439 869.52	0.13	195.375	0.207
5	1	4	6.5		4	4	1	5.5			439 882.28	0.12	195.375	0.318
5	1	4	3.5		4	4	1	2.5			439 888.17	0.14	195.375	0.167
1	1	1	1.5		0	0	0	1.5	451 713.550	0.100	451 713.54	0.03	0.000	0.333
1	1	1	2.5		0	0	0	1.5	451 715.773	0.100	451 715.73	0.03	0.000	0.500
1	1	1	0.5		0	0	0	1.5	451 717.454	0.100	451 717.39	0.03	0.000	0.167
5	2	3	3.5		6	1	6	4.5			462 245.82	0.47	227.757	0.182
5	2	3	6.5		6	1	6	7.5			462 249.81	0.44	227.756	0.308
5	2	3	4.5		6	1	6	5.5			462 262.36	0.46	227.756	0.217
5	2	3	5.5		6	1	6	6.5			462 266.29	0.45	227.756	0.259
8	3	5	6.5		9	2	8	7.5			470 655.46	1.87	551.558	0.206
8	3	5	9.5		9	2	8	10.5			470 657.24	1.87	551.558	0.289
8	3	5	7.5		9	2	8	8.5			470 666.40	1.87	551.558	0.231
8	3	5	8.5		9	2	8	9.5			470 668.15	1.87	551.558	0.259
9	3	7	9.5		8	4	4	8.5			471 785.92	1.67	608.168	0.259
9	3	7	8.5		8	4	4	7.5			471 787.48	1.67	608.168	0.231
9	3	7	10.5		8	4	4	9.5			471 795.15	1.67	608.168	0.289
9	3	7	7.5		8	4	4	6.5			471 796.78	1.67	608.168	0.206
8	4	5	6.5		9	1	8	7.5			475 930.00	1.84	551.558	0.206
8	4	5	9.5		9	1	8	10.5			475 931.78	1.84	551.558	0.289
8	4	5	7.5		9	1	8	8.5			475 940.97	1.84	551.558	0.231
8	4	5	8.5		9	1	8	9.5			475 942.72	1.84	551.558	0.259
6	5	1	4.5		6	4	2	4.5	488 697.500	0.700	488 696.98	0.08	375.246	0.184
6	5	1	7.5		6	4	2	7.5	488 697.500	0.700	488 697.56	0.08	375.246	0.299
6	5	1	5.5		6	4	2	5.5			488 700.08	0.08	375.246	0.210
6	5	1	6.5		6	4	2	6.5	488 700.700	0.200	488 700.77	0.08	375.246	0.249
5	3	3	3.5		6	0	6	4.5			492 652.59	0.47	227.756	0.182
5	3	3	6.5		6	0	6	7.5			492 656.66	0.44	227.756	0.308
5	3	3	4.5		6	0	6	5.5			492 669.55	0.46	227.756	0.217
5	3	3	5.5		6	0	6	6.5			492 673.59	0.45	227.756	0.259
2	2	1	0.5		2	1	2	0.5			504 230.04	0.04	38.264	0.050
2	2	1	1.5		2	1	2	0.5			504 238.27	0.04	38.264	0.050
2	2	1	0.5		2	1	2	1.5			504 240.36	0.03	38.263	0.050
2	2	1	3.5		2	1	2	3.5			504 243.25	0.04	38.264	0.343
2	2	1	1.5		2	1	2	1.5			504 248.59	0.03	38.263	0.080
2	2	1	2.5		2	1	2	3.5			504 251.39	0.04	38.264	0.057
2	2	1	3.5		2	1	2	2.5			504 253.69	0.04	38.263	0.057
2	2	1	2.5		2	1	2	1.5			504 254.50	0.04	38.263	0.070
2	2	1	1.5		2	1	2	2.5			504 255.92	0.04	38.263	0.070
2	2	1	2.5		2	1	2	2.5			504 261.83	0.03	38.263	0.173
6	2	5	6.5		5	3	2	5.5			532 413.85	0.31	263.612	0.259
6	2	5	5.5		5	3	2	4.5			532 417.01	0.31	263.612	0.217
6	2	5	7.5		5	3	2	6.5			532 426.20	0.31	263.612	0.308
6	2	5	4.5		5	3	2	3.5			532 429.45	0.32	263.612	0.182
7	5	2	5.5		7	4	3	5.5			554 586.17	0.14	486.981	0.193
7	5	2	8.5		7	4	3	8.5			554 586.60	0.14	486.981	0.293
7	5	2	6.5		7	4	3	6.5			554 588.25	0.14	486.981	0.218
7	5	2	7.5		7	4	3	7.5			554 588.67	0.14	486.981	0.251

TABLE II—*Continued*

J'	K'_a	K'_c	F'	←	J''	K''_a	K''_c	F''	ν_{exp}	$\Delta\nu_{exp}$	ν_{calc}	σ	E''	rel. Int.
3	3	1	1.5		3	2	2	1.5	565 784.428	0.100	565 784.42	0.02	96.302	0.114
3	3	1	4.5		3	2	2	4.5	565 789.475	0.100	565 789.43	0.03	96.302	0.327
3	3	1	2.5		3	2	2	2.5	565 795.348	0.100	565 795.34	0.02	96.302	0.147
3	3	1	3.5		3	2	2	3.5	565 800.436	0.100	565 800.41	0.02	96.302	0.218
6	4	2	4.5		6	3	3	4.5	567 951.013	0.600	567 950.67	0.10	356.301	0.184
6	4	2	7.5		6	3	3	7.5	567 951.013	0.600	567 951.36	0.10	356.301	0.299
6	4	2	5.5		6	3	3	5.5	567 953.476	0.600	567 953.28	0.10	356.301	0.210
6	4	2	6.5		6	3	3	6.5	567 953.476	0.600	567 953.91	0.10	356.301	0.249
5	5	0	3.5		5	4	1	3.5	573 023.677	0.300	573 023.87	0.05	277.135	0.170
5	5	0	6.5		5	4	1	6.5	573 025.156	0.150	573 025.17	0.05	277.135	0.306
5	5	0	4.5		5	4	1	4.5	573 029.277	0.150	573 029.42	0.05	277.135	0.199
5	5	0	5.5		5	4	1	5.5	573 030.884	0.150	573 030.91	0.04	277.135	0.244
8	6	2	6.5		8	5	3	6.5	589 776.765	0.300	589 776.64	0.10	635.104	0.201
8	6	2	9.5		8	5	3	9.5	589 776.765	0.300	589 776.92	0.10	635.104	0.289
8	6	2	7.5		8	5	3	7.5	589 778.759	0.300	589 778.58	0.10	635.104	0.223
8	6	2	8.5		8	5	3	8.5	589 778.759	0.300	589 778.91	0.10	635.104	0.253
5	3	2	3.5		5	2	3	3.5	612 666.100	0.200	612 666.24	0.05	243.175	0.170
5	3	2	6.5		5	2	3	6.5	612 667.500	0.200	612 667.44	0.05	243.175	0.306
5	3	2	4.5		5	2	3	4.5	612 670.070	0.200	612 670.12	0.05	243.175	0.199
5	3	2	5.5		5	2	3	5.5	612 671.330	0.200	612 671.24	0.05	243.175	0.244
7	6	1	5.5		7	5	2	5.5	618 923.100	0.400	618 922.90	0.11	505.480	0.193
7	6	1	8.5		7	5	2	8.5	618 923.100	0.400	618 923.32	0.11	505.480	0.293
7	6	1	6.5		7	5	2	6.5	618 926.300	0.400	618 926.05	0.11	505.480	0.218
7	6	1	7.5		7	5	2	7.5	618 926.300	0.400	618 926.62	0.11	505.480	0.251
4	4	1	2.5		4	3	2	3.5	646 863.317	0.100	646 863.35	0.03	173.798	0.018
4	4	1	2.5		4	3	2	2.5	646 867.719	0.100	646 867.71	0.03	173.797	0.149
4	4	1	5.5		4	3	2	5.5	646 870.372	0.100	646 870.34	0.03	173.798	0.315
4	4	1	3.5		4	3	2	3.5	646 875.574	0.100	646 875.58	0.02	173.798	0.181
4	4	1	4.5		4	3	2	4.5	646 878.279	0.100	646 878.30	0.02	173.798	0.236
4	4	1	4.5		4	3	2	5.5	646 882.382	0.100	646 882.41	0.03	173.798	0.018
8	2	6	8.5		7	5	3	7.5			657 426.55	1.05	492.942	0.259
8	2	6	7.5		7	5	3	6.5			657 428.53	1.05	492.942	0.228
8	2	6	9.5		7	5	3	8.5			657 437.00	1.05	492.942	0.294
8	2	6	6.5		7	5	3	5.5			657 438.97	1.05	492.942	0.200
4	2	2	2.5		4	1	3	2.5	666 124.339	0.100	666 124.39	0.03	148.027	0.149
4	2	2	5.5		4	1	3	5.5	666 126.787	0.100	666 126.77	0.03	148.027	0.315
4	2	2	3.5		4	1	3	3.5	666 130.944	0.100	666 130.89	0.03	148.027	0.181
4	2	2	4.5		4	1	3	4.5	666 133.242	0.100	666 133.20	0.03	148.027	0.236
9	7	2	7.5		9	6	3	7.5			682 096.32	1.25	800.900	0.206
9	7	2	10.5		9	6	3	10.5			682 096.48	1.25	800.900	0.285
9	7	2	8.5		9	6	3	8.5			682 098.32	1.25	800.900	0.227
9	7	2	9.5		9	6	3	9.5			682 098.59	1.25	800.900	0.254
2	0	2	2.5		1	1	1	2.5			687 152.96	0.04	15.068	0.090
2	0	2	2.5		1	1	1	1.5	687 155.068	0.300	687 155.15	0.04	15.068	0.210
2	0	2	1.5		1	1	1	0.5	687 158.857	0.300	687 159.00	0.04	15.068	0.083
2	0	2	1.5		1	1	1	1.5			687 162.85	0.04	15.068	0.107
2	0	2	3.5		1	1	1	2.5	687 163.764	0.300	687 163.93	0.04	15.068	0.400
2	0	2	0.5		1	1	1	0.5	687 169.705	0.300	687 169.84	0.04	15.068	0.083
2	0	2	0.5		1	1	1	1.5	687 173.554	0.300	687 173.68	0.04	15.068	0.017
3	1	2	1.5		3	0	3	1.5			708 457.02	0.05	71.367	0.114
3	1	2	4.5		3	0	3	4.5			708 462.68	0.05	71.367	0.327
3	1	2	2.5		3	0	3	2.5			708 468.96	0.04	71.366	0.147
3	1	2	3.5		3	0	3	3.5			708 474.55	0.04	71.366	0.218
10	7	3	8.5		10	6	4	8.5			732 036.49	3.59	960.415	0.211
10	7	3	11.5		10	6	4	11.5			732 036.70	3.60	960.416	0.282
10	7	3	9.5		10	6	4	9.5			732 037.89	3.60	960.416	0.230
10	7	3	10.5		10	6	4	10.5			732 038.09	3.60	960.416	0.254
2	1	2	2.5		1	0	1	2.5			735 114.58	0.04	13.742	0.090
2	1	2	1.5		1	0	1	0.5			735 115.50	0.04	13.743	0.083
2	1	2	2.5		1	0	1	1.5	735 122.772	0.300	735 122.88	0.04	13.742	0.210
2	1	2	3.5		1	0	1	2.5	735 125.136	0.300	735 125.02	0.04	13.742	0.400
2	1	2	0.5		1	0	1	0.5			735 125.82	0.04	13.743	0.083
2	1	2	1.5		1	0	1	1.5			735 130.21	0.04	13.742	0.107
6	6	0	4.5		6	5	1	4.5			739 666.57	0.11	391.547	0.184
6	6	0	7.5		6	5	1	7.5			739 667.50	0.10	391.547	0.299
6	6	0	5.5		6	5	1	5.5			739 671.72	0.10	391.547	0.210
6	6	0	6.5		6	5	1	6.5			739 672.85	0.10	391.548	0.249
5	5	1	3.5		5	4	2	3.5			744 456.60	0.05	270.829	0.170
5	5	1	6.5		5	4	2	6.5			744 458.21	0.06	270.829	0.306
5	5	1	4.5		5	4	2	4.5			744 462.91	0.05	270.829	0.199
5	5	1	5.5		5	4	2	5.5			744 464.65	0.05	270.829	0.244
3	2	2	1.5		3	1	3	1.5			746 370.84	0.05	71.406	0.114
3	2	2	4.5		3	1	3	4.5			746 376.98	0.05	71.406	0.327
3	2	2	2.5		3	1	3	2.5			746 383.94	0.04	71.406	0.147
3	2	2	3.5		3	1	3	3.5			746 390.06	0.04	71.406	0.218
9	6	3	7.5		9	5	4	7.5			748 491.83	1.93	775.933	0.206
9	6	3	10.5		9	5	4	10.5			748 492.17	1.93	775.933	0.285
9	6	3	8.5		9	5	4	8.5			748 493.42	1.93	775.933	0.227
9	6	3	9.5		9	5	4	9.5			748 493.70	1.93	775.933	0.254

TABLE II—*Continued*

J'	K'_a	K'_c	F'	←	J''	K''_a	K''_c	F''	ν_{exp}	$\Delta\nu_{exp}$	ν_{calc}	σ	E''	rel. Int.
4	3	2	2.5		4	2	3	2.5			764 530.84	0.04	148.295	0.149
4	3	2	5.5		4	2	3	5.5			764 533.72	0.04	148.295	0.315
4	3	2	3.5		4	2	3	3.5			764 539.06	0.04	148.295	0.181
4	3	2	4.5		4	2	3	4.5			764 541.95	0.04	148.295	0.236
9	3	6	7.5		10	2	9	8.5			767 288.67	3.59	660.351	0.211
9	3	6	10.5		10	2	9	11.5			767 290.14	3.59	660.351	0.286
9	3	6	8.5		10	2	9	9.5			767 298.83	3.59	660.350	0.233
9	3	6	9.5		10	2	9	10.5			767 300.27	3.59	660.350	0.258
9	4	6	7.5		10	1	9	8.5			768 445.59	3.55	660.351	0.211
9	4	6	10.5		10	1	9	11.5			768 447.06	3.55	660.351	0.286
9	4	6	8.5		10	1	9	9.5			768 455.75	3.55	660.350	0.233
9	4	6	9.5		10	1	9	10.5			768 457.19	3.55	660.350	0.258
6	2	4	4.5		7	1	7	5.5			777 789.22	1.02	298.722	0.192
6	2	4	7.5		7	1	7	8.5			777 792.25	1.00	298.722	0.300
6	2	4	5.5		7	1	7	6.5			777 804.12	1.01	298.722	0.223
6	2	4	6.5		7	1	7	7.5			777 807.11	1.00	298.722	0.259
8	7	1	6.5		8	6	2	6.5			784 034.14	0.50	654.777	0.201
8	7	1	9.5		8	6	2	9.5			784 034.44	0.50	654.777	0.289
8	7	1	7.5		8	6	2	7.5			784 037.32	0.50	654.777	0.223
8	7	1	8.5		8	6	2	8.5			784 037.81	0.50	654.777	0.253
6	3	4	4.5		7	0	7	5.5			784 350.16	1.02	298.722	0.192
6	3	4	7.5		7	0	7	8.5			784 353.19	1.00	298.722	0.300
6	3	4	5.5		7	0	7	6.5			784 365.13	1.01	298.722	0.223
6	3	4	6.5		7	0	7	7.5			784 368.13	1.01	298.722	0.259
5	4	2	3.5		5	3	3	3.5			798 617.86	0.07	244.190	0.170
5	4	2	6.5		5	3	3	6.5			798 619.49	0.07	244.190	0.306
5	4	2	4.5		5	3	3	4.5			798 623.78	0.07	244.190	0.199
5	4	2	5.5		5	3	3	5.5			798 625.44	0.07	244.190	0.244
8	5	3	6.5		8	4	4	6.5			807 512.88	0.63	608.168	0.201
8	5	3	9.5		8	4	4	9.5			807 513.37	0.63	608.168	0.289
8	5	3	7.5		8	4	4	7.5			807 514.95	0.63	608.168	0.223
8	5	3	8.5		8	4	4	8.5			807 515.36	0.63	608.168	0.253
5	2	4	5.5		4	3	1	4.5			826 104.34	0.12	182.537	0.258
5	2	4	4.5		4	3	1	3.5			826 108.40	0.13	182.537	0.207
5	2	4	6.5		4	3	1	5.5			826 117.13	0.12	182.537	0.318
5	2	4	3.5		4	3	1	2.5			826 121.22	0.14	182.537	0.167
10	8	2	8.5		10	7	3	8.5			828 323.95	5.85	984.834	0.211
10	8	2	11.5		10	7	3	11.5			828 324.01	5.85	984.834	0.282
10	8	2	9.5		10	7	3	9.5			828 326.06	5.85	984.834	0.230
10	8	2	10.5		10	7	3	10.5			828 326.30	5.85	984.834	0.254
7	2	5	7.5		6	5	2	6.5			829 720.44	0.62	387.468	0.259
7	2	5	6.5		6	5	2	5.5			829 723.07	0.62	387.468	0.223
7	2	5	8.5		6	5	2	7.5			829 732.36	0.62	387.468	0.300
7	2	5	5.5		6	5	2	4.5			829 734.90	0.62	387.468	0.192
8	3	6	8.5		7	4	3	7.5			836 359.32	1.00	486.981	0.259
8	3	6	7.5		7	4	3	6.5			836 361.10	1.00	486.981	0.228
8	3	6	9.5		7	4	3	8.5			836 368.45	1.01	486.981	0.294
8	3	6	6.5		7	4	3	5.5			836 370.33	1.00	486.981	0.200
6	5	2	4.5		6	4	3	4.5			851 542.48	0.11	359.064	0.184
6	5	2	7.5		6	4	3	7.5			851 543.51	0.11	359.064	0.299
6	5	2	5.5		6	4	3	5.5			851 547.18	0.11	359.064	0.210
6	5	2	6.5		6	4	3	6.5			851 548.27	0.11	359.064	0.249
6	6	1	4.5		6	5	2	4.5			853 572.39	0.12	387.468	0.184
6	6	1	7.5		6	5	2	7.5			853 573.46	0.12	387.468	0.299
6	6	1	5.5		6	5	2	5.5			853 577.77	0.11	387.468	0.210
6	6	1	6.5		6	5	2	6.5			853 578.98	0.11	387.468	0.249
6	2	4	6.5		5	5	1	5.5			869 542.97	0.31	295.662	0.259
6	2	4	5.5		5	5	1	4.5			869 546.98	0.31	295.661	0.217
6	2	4	7.5		5	5	1	6.5			869 558.00	0.31	295.661	0.308
6	2	4	4.5		5	5	1	3.5			869 561.79	0.32	295.661	0.182
7	4	3	5.5		7	3	4	5.5	882 363.799	0.500	882 363.48	0.15	457.549	0.193
7	4	3	8.5		7	3	4	8.5	882 363.799	0.500	882 364.22	0.15	457.549	0.293
7	4	3	6.5		7	3	4	6.5	882 366.859	0.500	882 366.47	0.15	457.549	0.218
7	4	3	7.5		7	3	4	7.5	882 366.859	0.500	882 367.11	0.15	457.549	0.251
7	7	0	5.5		7	6	1	5.5	900 791.915	0.500	900 791.43	0.15	526.125	0.193
7	7	0	8.5		7	6	1	8.5	900 791.915	0.500	900 792.10	0.15	526.125	0.293
7	7	0	6.5		7	6	1	6.5	900 796.655	0.500	900 796.13	0.14	526.126	0.218
7	7	0	7.5		7	6	1	7.5	900 796.655	0.500	900 797.00	0.14	526.126	0.251
7	6	2	5.5		7	5	3	5.5	923 725.943	0.500	923 725.52	0.12	492.942	0.193
7	6	2	8.5		7	5	3	8.5	923 725.943	0.500	923 726.22	0.12	492.942	0.293
7	6	2	6.5		7	5	3	6.5	923 729.950	0.500	923 729.51	0.12	492.942	0.218
7	6	2	7.5		7	5	3	7.5	923 729.950	0.500	923 730.29	0.12	492.942	0.251
4	2	3	4.5		3	3	0	3.5	930 741.958	0.100	930 741.93	0.04	117.249	0.255
4	2	3	3.5		3	3	0	2.5	930 748.783	0.300	930 748.67	0.05	117.249	0.191
4	2	3	5.5		3	3	0	4.5	930 757.496	0.100	930 757.51	0.05	117.249	0.333
4	2	3	2.5		3	3	0	1.5	930 764.192	0.300	930 764.17	0.06	117.249	0.143
6	3	3	4.5		6	2	4	4.5			948 384.85	0.09	324.666	0.184
6	3	3	7.5		6	2	4	7.5	948 385.713	1.000	948 386.01	0.09	324.666	0.299
6	3	3	5.5		6	2	4	5.5	948 390.014	1.000	948 389.30	0.08	324.666	0.210
6	3	3	6.5		6	2	4	6.5	948 390.014	1.000	948 390.39	0.08	324.666	0.249

TABLE II—*Continued*

J'	K'_a	K'_c	F'	←	J''	K''_a	K''_c	F''	ν_{exp}	$\Delta\nu_{exp}$	ν_{calc}	σ	E''	rel. Int.
9	8	1	7.5		9	7	2	7.5			961 888.65	1.71	823.652	0.206
9	8	1	10.5		9	7	2	10.5			961 888.86	1.71	823.652	0.285
9	8	1	8.5		9	7	2	8.5			961 891.77	1.71	823.652	0.227
9	8	1	9.5		9	7	2	9.5			961 892.18	1.71	823.652	0.254
7	7	1	5.5		7	6	2	5.5			968 033.68	0.17	523.754	0.193
7	7	1	8.5		7	6	2	8.5			968 034.42	0.17	523.754	0.293
7	7	1	6.5		7	6	2	6.5			968 038.42	0.17	523.754	0.218
7	7	1	7.5		7	6	2	7.5			968 039.32	0.17	523.754	0.251
3	0	3	2.5		2	1	2	1.5	992 398.366	1.000	992 397.66	0.03	38.263	0.160
3	0	3	3.5		2	1	2	2.5	992 398.366	1.000	992 398.95	0.03	38.263	0.245
3	0	3	1.5		2	1	2	0.5			992 400.41	0.04	38.264	0.100
3	0	3	4.5		2	1	2	3.5			992 401.76	0.03	38.264	0.357
3	0	3	2.5		2	1	2	2.5			992 405.00	0.04	38.263	0.052
5	2	3	3.5		5	1	4	3.5	993 142.663	0.300	993 142.82	0.06	210.048	0.170
5	2	3	6.5		5	1	4	6.5	993 144.874	0.300	993 144.80	0.06	210.048	0.306
5	2	3	4.5		5	1	4	4.5	993 149.455	0.300	993 149.56	0.06	210.048	0.199
5	2	3	5.5		5	1	4	5.5	993 151.536	0.300	993 151.48	0.06	210.047	0.244
10	3	7	10.5		9	6	4	9.5			994 000.20	3.44	780.648	0.258
10	3	7	9.5		9	6	4	8.5			994 001.42	3.44	780.648	0.233
10	3	7	11.5		9	6	4	10.5			994 008.22	3.44	780.648	0.286
10	3	7	8.5		9	6	4	7.5			994 009.44	3.44	780.648	0.211
3	1	3	2.5		2	0	2	1.5	1 001 814.085	0.100	1 001 814.14	0.03	37.989	0.160
3	1	3	3.5		2	0	2	2.5	1 001 815.761	0.100	1 001 815.82	0.03	37.989	0.245
3	1	3	1.5		2	0	2	0.5			1 001 816.33	0.03	37.989	0.100
3	1	3	4.5		2	0	2	3.5	1 001 818.136	0.100	1 001 818.04	0.03	37.989	0.357
3	1	3	2.5		2	0	2	2.5	1 001 821.979	0.300	1 001 821.84	0.04	37.989	0.052
8	7	2	6.5		8	6	3	6.5	1 012 867.431	0.300	1 012 867.26	0.10	645.879	0.201
8	7	2	9.5		8	6	3	9.5	1 012 867.431	0.300	1 012 867.74	0.10	645.879	0.289
8	7	2	7.5		8	6	3	7.5	1 012 871.162	0.300	1 012 870.80	0.10	645.879	0.223
8	7	2	8.5		8	6	3	8.5	1 012 871.162	0.300	1 012 871.39	0.10	645.879	0.253
4	1	3	2.5		4	0	4	2.5	1 017 762.140	0.300	1 017 762.26	0.05	114.078	0.149
4	1	3	5.5		4	0	4	5.5	1 017 765.983	0.100	1 017 766.01	0.04	114.078	0.315
4	1	3	3.5		4	0	4	3.5	1 017 772.625	0.300	1 017 772.76	0.04	114.078	0.181
4	1	3	4.5		4	0	4	4.5	1 017 776.505	0.100	1 017 776.46	0.04	114.078	0.236
5	3	3	3.5		5	2	4	3.5	1 022 183.611	0.300	1 022 183.81	0.04	210.093	0.170
5	3	3	6.5		5	2	4	6.5	1 022 185.884	0.100	1 022 185.85	0.04	210.093	0.306
5	3	3	4.5		5	2	4	4.5	1 022 190.916	0.100	1 022 190.94	0.04	210.093	0.199
5	3	3	5.5		5	2	4	5.5	1 022 192.993	0.100	1 022 192.96	0.04	210.093	0.244
6	4	3	4.5		6	3	4	4.5			1 024 638.93	0.09	324.885	0.184
6	4	3	7.5		6	3	4	7.5	1 024 639.799	0.500	1 024 640.17	0.09	324.885	0.299
6	4	3	5.5		6	3	4	5.5	1 024 644.778	0.500	1 024 644.10	0.09	324.885	0.210
6	4	3	6.5		6	3	4	6.5	1 024 644.778	0.500	1 024 645.33	0.09	324.885	0.249
4	2	3	2.5		4	1	4	2.5	1 025 646.141	0.300	1 025 646.29	0.05	114.084	0.149
4	2	3	5.5		4	1	4	5.5	1 025 650.059	0.100	1 025 650.08	0.04	114.083	0.315
4	2	3	3.5		4	1	4	3.5			1 025 656.96	0.04	114.083	0.181
4	2	3	4.5		4	1	4	4.5	1 025 660.744	0.100	1 025 660.72	0.04	114.083	0.236
7	5	3	5.5		7	4	4	5.5	1 038 370.740	1.000	1 038 370.47	0.17	458.305	0.193
7	5	3	8.5		7	4	4	8.5	1 038 370.740	1.000	1 038 371.28	0.17	458.305	0.293
7	5	3	6.5		7	4	4	6.5	1 038 374.823	1.000	1 038 374.48	0.17	458.305	0.218
7	5	3	7.5		7	4	4	7.5	1 038 374.823	1.000	1 038 375.30	0.17	458.305	0.251
8	8	0	6.5		8	7	1	6.5	1 045 395.276	0.400	1 045 395.06	0.13	680.930	0.201
8	8	0	9.5		8	7	1	9.5	1 045 395.276	0.400	1 045 395.54	0.14	680.930	0.289
8	8	0	7.5		8	7	1	7.5	1 045 399.643	0.400	1 045 399.32	0.13	680.930	0.223
8	8	0	8.5		8	7	1	8.5	1 045 399.643	0.400	1 045 400.00	0.13	680.930	0.253
10	6	4	8.5		10	5	5	8.5			1 067 474.86	5.19	924.808	0.211
10	6	4	11.5		10	5	5	11.5			1 067 475.28	5.19	924.808	0.282
10	6	4	9.5		10	5	5	9.5			1 067 476.68	5.19	924.808	0.230
10	6	4	10.5		10	5	5	10.5			1 067 476.99	5.19	924.808	0.254
8	6	3	6.5		8	5	4	6.5	1 068 331.810	0.600	1 068 331.55	0.19	610.243	0.201
8	6	3	9.5		8	5	4	9.5	1 068 331.810	0.600	1 068 332.10	0.19	610.243	0.289
8	6	3	7.5		8	5	4	7.5	1 068 335.298	0.600	1 068 334.86	0.18	610.243	0.223
8	6	3	8.5		8	5	4	8.5	1 068 335.298	0.600	1 068 335.44	0.18	610.243	0.253
2	2	1	1.5		1	1	0	1.5	1 071 040.940	0.300	1 071 040.98	0.04	19.357	0.107
2	2	1	2.5		1	1	0	1.5	1 071 046.625	0.300	1 071 046.88	0.04	19.357	0.210
2	2	1	3.5		1	1	0	2.5	1 071 049.056	0.300	1 071 049.02	0.04	19.357	0.400
2	2	1	0.5		1	1	0	0.5			1 071 051.43	0.04	19.357	0.083
2	2	1	2.5		1	1	0	2.5	1 071 057.016	0.300	1 071 057.16	0.04	19.357	0.090
2	2	1	1.5		1	1	0	0.5	1 071 059.808	0.300	1 071 059.66	0.04	19.357	0.083
7	2	5	5.5		8	1	8	6.5			1 080 805.16	2.05	379.093	0.200
7	2	5	8.5		8	1	8	9.5			1 080 807.51	2.03	379.093	0.294
7	2	5	6.5		8	1	8	7.5			1 080 818.57	2.04	379.093	0.228
7	2	5	7.5		8	1	8	8.5			1 080 820.89	2.04	379.093	0.259
8	8	1	6.5		8	7	2	6.5			1 081 588.78	0.24	679.664	0.201
8	8	1	9.5		8	7	2	9.5			1 081 589.30	0.24	679.664	0.289
8	8	1	7.5		8	7	2	7.5			1 081 593.03	0.24	679.664	0.223
8	8	1	8.5		8	7	2	8.5			1 081 593.72	0.24	679.665	0.253
7	3	5	5.5		8	0	8	6.5			1 082 079.64	2.05	379.093	0.200
7	3	5	8.5		8	0	8	9.5			1 082 082.00	2.04	379.093	0.294
7	3	5	6.5		8	0	8	7.5			1 082 093.06	2.05	379.093	0.228
7	3	5	7.5		8	0	8	8.5			1 082 095.38	2.04	379.093	0.259

TABLE III

$H_2{}^{36}S$: Observed Submillimeter Transition Frequencies in MHz

J'	K'_a	K'_c	$\leftarrow$	J''	K''_a	K''_c	ν_{exp}	$\Delta\nu_{exp}$[a]
3	3	1		3	2	2	559250.950	0.100
4	4	1		4	3	2	636677.520	0.100
2	0	2		1	1	1	686766.635	0.100

Note. Frequencies given in MHz.
[a] Estimated measurement uncertainty.

high-frequency backward wave oscillators supplied by the ISTOK Research and Production Co., located in Fryazino, near Moscow, Russia, and digitally phase locked to a commercially available 78-118 GHz synthesizer from the Institute of Electronic Measurements, KVARZ, Nizhnii Novgorod, Russia. In this configuration, we have achieved the first measurements with phase-locked BWOs beyond 1 THz (*1*). All measurements were carried out in natural abundance, i.e., 4.2% for $H_2{}^{34}S$, 0.76% for $H_2{}^{33}S$, and 0.02% for $H_2{}^{36}S$. The spectra were recorded at pressures between 15 and 40 μbar. The absorption cell was 4.5 m long. The signal was detected by a liquid-He-cooled InSb detector. The measured frequencies of $H_2{}^{34}S$, $H_2{}^{33}S$, and $H_2{}^{36}S$ are listed in Tables I, II, and III, respectively. Figures 1 to 3 display a few sample spectra.

III. HAMILTONIAN

All observable effects are included in the appropriate Hamiltonian

$$H = H_{\mathrm{Rot}} + H_{\mathrm{Q}} + H_{\mathrm{NSR}}, \tag{1}$$

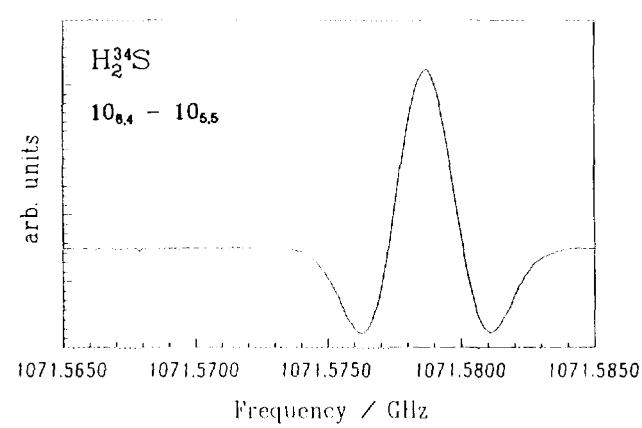

FIG. 1. The observed spectrum of $H_2{}^{34}S$ at 1071 GHz. The spectrum was recorded in the second derivative form.

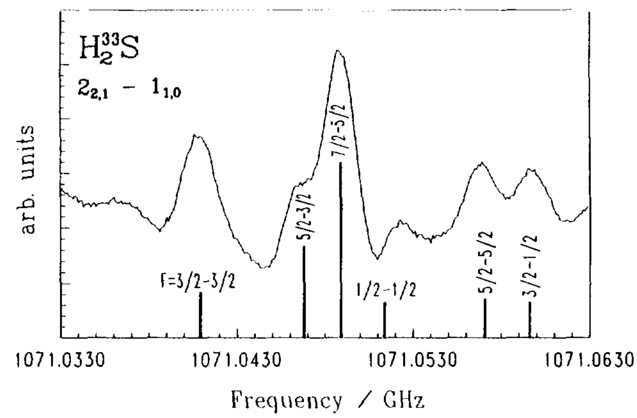

FIG. 2. The resolved hyperfine structure of $H_2{}^{33}S$ at 1071 GHz. The calculated line positions and relative intensities are indicated.

where H_{Rot} is the rotational part, H_Q denotes the nuclear electric quadrupole, and H_{NSR} denotes the nuclear spin–rotation interaction Hamiltonians.

For H_{Rot}, the flexibility of the S–H bonds as well as the asymmetry ($\kappa \sim 0.53$) causes the calculation of the rotational spectra to be remarkably sensitive to the choice of the reduced Hamiltonian and the representation. This has been pointed out by Strow (*14*) after fitting rotational and rovibrational H_2S transitions to different types of Watson-type reduced Hamiltonians. Although the asymmetry parameter indicates an oblate rather than a prolate top, the A-reduction in III^r representation fails to fit the experimental data satisfactorily, while the best fits have been achieved with an A-type reduced Hamiltonian in I^r representation. On the basis of a larger set of rotational transitions, Yamada and Klee (*15*) have recently reported that comparably good fits can also be achieved when using the S-reduction in both I^r and III^r representations. Burenin *et al.* (*16*) chose a Padé-type Hamiltonian and showed that the reproduction of the observed spectra is superior to the Watson-type approach with the same number of variable parameters. Nevertheless, in this paper we have employed the more com-

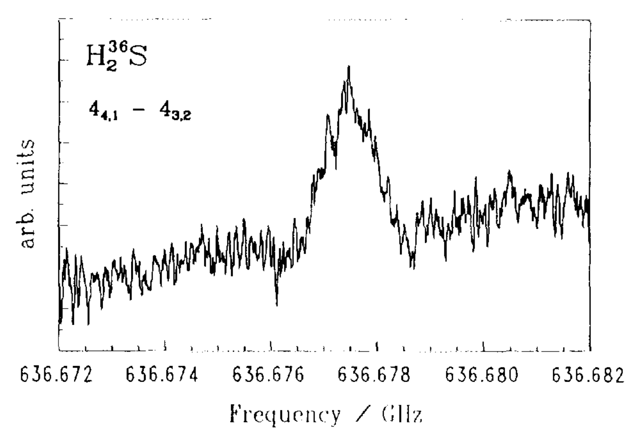

FIG. 3. One of the three weak lines observed between 550 and 700 GHz and tentatively assigned to $H_2{}^{36}S$.

TABLE IV

Rotational Constants in MHz of $H_2{}^{33}S$ and $H_2{}^{34}S$ Determined in the Present Work, in Comparison to the Values from Ref. (6)

Parameter	$H_2{}^{33}S$ Ref. (6)	$H_2{}^{33}S$ here	$H_2{}^{34}S$ here	$H_2{}^{34}S$ Ref. (6)
A	310031.115 (276)	310032.4151(156)[a]	309509.1803 (110)	309508.381 (177)
B	270356.133 (360)	270356.5930 (229)	270356.0827 (122)	270355.496 (234)
C	141706.807 (219)	141707.9187 (178)	141597.4210 (112)	141596.584 (168)
Δ_J	19.56158 (201)	19.57159 (104)	19.56956 (62)	19.55867 (257)
Δ_{JK}	-68.3552 (78)	-68.3324 (24)	-68.29519 (160)	-68.3133 (93)
Δ_K	110.7872 (69)	110.7785 (21)	110.53695 (153)	110.5370 (101)
δ_J	8.86079 (96)	8.86103 (58)	8.861592 (185)	8.86095 (129)
δ_K	-4.0341 (39)	-4.02557 (156)	-4.08378 (107)	-4.0929 (45)
$H_J \times 10^3$	8.1290[b]	8.265 (25)	8.2645 (109)	8.1342 (159)
$H_{JK} \times 10^3$	-45.999[b]	-46.076 (135)	-46.033 (58)	-46.043 (96)
$H_{KJ} \times 10^3$	37.818[b]	37.77 (28)	38.380 (104)	37.886 (218)
$H_K \times 10^3$	41.194[b]	41.52 (20)	40.811 (66)	40.983 (144)
$h_J \times 10^3$	4.0632[b]	4.0983 (126)	4.1092 (47)	4.0669 (81)
$h_{KJ} \times 10^3$	-14.562[b]	-14.661 (90)	-14.6780 (36)	-14.582 (39)
$h_K \times 10^3$	36.717[b]	37.565 (159)	37.171 (53)	36.590 (51)
$L_J \times 10^6$	-4.182[b]	-4.862 (182)	-4.848 (59)	-4.182[b]
$L_{KJJ} \times 10^6$	35.51[b]	36.30 (119)	37.86 (56)	35.51[b]
$L_{KJ} \times 10^6$	-99.52[b]	-98.6 (53)	-105.62 (192)	-99.52[b]
$L_{KKJ} \times 10^6$	164.3[b]	160.2 (92)	160.0 (29)	164.3[b]
$L_K \times 10^6$	-134.3[b]	-133.8 (54)	-129.27 (159)	-134.3[b]
$l_J \times 10^6$	-2.112[b]	-2.394 (89)	-2.414 (29)	-2.112[b]
$l_{JK} \times 10^6$	12.15[b]	17.73 (148)	14.73 (29)	12.15[b]
$l_{KJ} \times 10^6$	-9.04[b]	-26.30 (180)	-19.06 (155)	-9.04[b]
$l_K \times 10^6$	-52.70[b]	-46.6 (48)	-46.92 (161)	-52.70[b]
$P_{KKJ} \times 10^9$	10.85[b]	0.0	0.0	10.85[b]
$P_{KKKJ} \times 10^9$	-94.25[b]	0.0	0.0	-94.25[b]
$P_K \times 10^9$	110.3[b]	33.0 (77)	37.52 (144)	110.3[b]
$p_{KJJ} \times 10^9$	0.0	-31.3 (62)	-8.04 (80)	0.0
$p_{KKJ} \times 10^9$	-57.92[b]	0.0	-12 (16)	-57.92[b]
$p_K \times 10^9$	120.5[b]	56(42)	64.2 (189)	120.5[b]

[a] Values in brackets: 1σ. Parameter fixed when no errors are given.
[b] Value for $H_2{}^{32}S$ from Ref. (6).

monly used A-reduced Watsonian in the I^r representation as first suggested by Strow. Here, the definition of rotational and centrifugal constants is consistent with that of Flaud *et al.* (6). It should, however, be pointed out that the sign convention for the higher terms in extending the Watson A-reduced Hamiltonian has to be considered. Flaud *et al.* (6) chose all terms with positive signs, whereas Yamada and Klee (15) have chosen the signs of the higher terms following the traditional convention of an alternative power series. According to this convention, the J^8 (L terms) have negative signs and the J^{10} are positive. Therefore attention must be paid to the signs of the L term in comparison with other papers. For H_Q and H_{NSR}, due to the ^{33}S nucleus, the appropriate Hamiltonian is given in the literature (see e.g., Refs. 17, 18).

Relative intensities s of $H_2{}^{33}S$ hyperfine splittings are calculated by the usual expression (17):

$$s(J'IF' \leftarrow J''IF'') = \frac{(2F'+1)(2F''+1)}{2I+1} \begin{Bmatrix} J' & F' & I \\ F'' & J' & I \end{Bmatrix}^2 . \qquad (2)$$

IV. ANALYSIS

For $H_2{}^{34}S$ and $H_2{}^{33}S$, the transition frequencies, together with the FIR data from Ref. (*6*), have been fitted in a least-squares procedure to the Hamiltonian discussed above. Table IV shows the rotational constants derived in the present study, in comparison with previous values by Flaud *et al.* (*6*). Quartic, sextic, and octic centrifugal distortion constants have also been determined. To obtain an estimate for the model error of using an A-reduced Hamiltonian, the present $H_2{}^{34}S$ data set has also been fitted to a S-reduced Hamiltonian in I^r representation. The resulting predicted frequencies deviate from those given in Table I by 5σ or less. Since only 3 lines of $H_2{}^{36}S$ have been identified tentatively, precise molecular constants could not be derived for this rare isotopomer.

In the case of $H_2{}^{33}S$, the electric quadrupole hyperfine constants for the ^{33}S nucleus were determined. Hyperfine matrix elements not diagonal in J have been found not to improve the fit and have subsequently been neglected.

The analysis of the $H_2{}^{33}S$ spectra also revealed the small shifts caused by the nuclear spin–rotation interaction. The ^{33}S hyperfine parameters are given in Table V. We may note that when dividing the constants by the corresponding rotational constants, their order of magnitude is consistent with their equivalents for C ^{33}S (*19*) and HS ^{33}SH (*18*).

V. CONCLUSION

The new measurements near 1 THz of the pure rotational transitions of $H_2{}^{34}S$ and $H_2{}^{33}S$ and their fit to the appropriate Hamiltonian allow highly accurate frequency

TABLE V

$H_2{}^{33}S$: Hyperfine Constants in MHz for the ^{33}S Nucleus in Comparison to Previous Values from Ref. (*2*)

Parameter	here	Ref. (*2*)
eQq_{aa}	-32.820 (53)[a]	-32
$eQq_{bb} - eQq_{cc}$	-50.013 (111)	
eQq_{bb}	-8.597[b] (66)	-8
eQq_{cc}	41.416[b] (56)	40
$C_{aa} + C_{bb} + C_{cc}$	0.0825 (147)	
$C_{bb} - C_{cc}$	0.0298 (115)	
$2C_{aa} - C_{bb} - C_{cc}$	-0.0573 (140)	
C_{aa}	0.0084[b] (72)	
C_{bb}	0.0520[b] (86)	
C_{cc}	0.0222[b] (72)	

[a] Values in brackets: 1σ.
[b] Derived value.

predictions for these two species, which are of astrophysical relevance. Furthermore, the rare $H_2{}^{36}S$ isotopomer has been detected for the first time. The hyperfine parameters determined for the ^{33}S nucleus may serve as definite tests to quantum chemical ab initio calculations, yielding information about the role of hybridization involving $3d$ orbitals since the electric quadrupole interaction constants reflect the electronic distribution near the sulfur nucleus.

ACKNOWLEDGMENTS

The authors thank Dr. M. Liedtke and M. Yu. Tretyakov for help with the measurements in the submillimeter-wave region and A. Dolgner for the sample preparation. A.H.S. thanks Dr. J. Demaison for helpful discussion. M.T. acknowledges financial support of Morino Foundation for Molecular Science and the Deutsche Forschungsgemeinschaft (DFG) for the travel expenses to and the stay at Köln. This work has been supported in part by the DFG via Grant SFB-301.

RECEIVED: November 2, 1994

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