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The Centrifugal Distortion Constants of Disulfur Monoxide

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The microwave spectrum of the molecular transient disulfur monoxide, S_2O, has been reexamined and the microwave measurements have been extended into the millimeterwave region. From the present data, the following ground-state rotational constants and quartic distortion constants have been obtained (MHz):

$$A = 41\,915.44,\quad B = 5059.07,\quad C = 4507.19$$
$$\Delta_J = 1.895 \times 10^{-3},\quad \Delta_{JK} = -3.192 \times 10^{-2},\quad \Delta_K = 1.197 \times 10^{0},$$
$$\delta_J = 3.453 \times 10^{-4},\ \text{and}\quad \delta_K = 1.223 \times 10^{-2}.$$

The centrifugal distortion constants obtained from the rotational spectrum are used to discuss the vibrational spectrum of disulfur monoxide.

I. INTRODUCTION

The first unambiguous spectroscopic evidence for the existence of the molecular transient disulfur monoxide, SSO, was obtained from the discharge studies of Meschi and Myers (*1*). They reported the first measurements of that molecule in the microwave region and were able to observe enough transitions of the various isotopes to obtain the molecular structure of the bent S_2O molecule.

The study presented here was undertaken to obtain an improved and more complete set of rotational parameters with the aim to use the measured distortion constants to obtain information on the fundamental vibrational frequencies, which have not yet been clearly established. Furthermore, we wished to study further the question of what is the major constituent in a microwave discharge of a mixture of SO_2 and sulfur vapor. Since the initial studies by Schenk (*2*), this problem has been studied by a number of workers with controversial results (*3–5*). Various sulfur oxides, such as SO, and S_2O_2, were put forward to be the main constituent in the discharge product. In order to ensure the unequivocal presence of S_2O, we have employed various preparation schemes.

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Sufficient rotational transitions in the microwave and millimeter wave region have now been measured to evaluate the centrifugal distortion constants of disulfur monoxide.

II. EXPERIMENTAL

A. Preparation of Disulfur Monoxide

The S_2O molecule was prepared by two methods: by a microwave discharge burning in SO_2 mixed with sulfur vapor and by chemical preparation.

In the discharge preparation scheme of S_2O, it has been possible to use the millimeter-wave spectrometer to study the conditions for the preparation of S_2O and also SO. This was made possible by monitoring the known rotational spectra of the discharge products S_2O and SO, while changing the composition of the carrier gases SO_2 and sulfur vapor in the discharge tube. Our observations do not indicate whether S_2O_2 was present.

Disulfur monoxide was obtained as a discharge product only with an abundance of sulfur vapor being present (obtained by heating sulfur powder to about 70°C) while passing SO_2 through the discharge tube. A reduction in sulfur vapor yielded a sharp decrease in the production rate of S_2O. With no sulfur vapor being present, the radical SO was being produced as reported earlier by Winnewisser *et al.* (*6*).

All of the S_2O rotational lines obtained in this way were compared with chemically prepared samples according to methods described by Schenk *et al.* (*7*). At temperatures between 140 and 160°C thionylchloride was reacted with various metal sulfides, such as CdS, PbS, and CuS, according to the equation

$$SOCl_2 + CdS \rightarrow S_2O + CdCl_2.$$

It was found that the S_2O sample was always contaminated with about 20% SO_2. Furthermore, the disulfur monoxide lasted only for a few minutes in the free space absorption cell before decomposition into SO_2, and elemental sulfur became noticeable.

B. Measurement of the Rotational Spectrum

The microwave measurements were carried out with a parallel plate Stark modulated spectrometer described by Winnewisser and Cook (*8*). In addition, an ordinary X-band Stark cell was also employed. Disulfur monoxide had the shortest lifetime in this cell. The millimeterwave measurements were obtained using the spectrometer described elsewhere (*9*). The millimeterwave frequencies were generated by harmonic multiplication of the fundamental frequencies. Most of the lines were measured using chemically prepared samples of S_2O, and all lines were measured with the cell at room temperature. The observed rotational transitions are collected in Table I.

III. ANALYSIS

The rotational Hamiltonian including the effects of centrifugal distortion can be written in the form (*10*)

$$\begin{aligned}\mathcal{H} = {} & \tfrac{1}{2}(\mathcal{B} + \mathcal{C})P^2 + [\mathcal{A} - \tfrac{1}{2}(\mathcal{B} + \mathcal{C})]\{P_z^2 - b_p P_-^2\} \\ & - \Delta_J P^4 - \Delta_{JK} P^2 P_z^2 - \Delta_K P_z^4 - 2\delta_J P^2 P_-^2 - \delta_K [P_z^2 P_-^2 + P_-^2 P_z^2], \quad (1)\end{aligned}$$

where $P^2 = P_x^2 + P_y^2 + P_z^2$, $P_-^2 = P_x^2 - P_y^2$ with $z = a$, $x = b$, $y = c$, and

TABLE I

ROTATIONAL TRANSITIONS OF S_2O (MHz)

Transition	Observed Frequency	Calculated Frequency[a]	Deviation[b]	Ref.
$1_{1,0} \leftarrow 1_{0,1}$	37 407.2	37 407.12	0.08	c
$1_{1,1} \leftarrow 0_{0,0}$	46 421.6	46 421.49	0.11	c
$2_{0,2} \leftarrow 1_{0,1}$	19 126.4	19 126.32	0.08	c
$2_{1,1} \leftarrow 1_{1,0}$	19 684.3	19 684.47	-0.17	c
$2_{1,1} \leftarrow 2_{0,2}$	37 965.3	37 965.28	0.02	c
$3_{0,3} \leftarrow 2_{0,2}$	28 673.78	28 673.99	-0.21	d
$3_{1,3} \leftarrow 2_{1,2}$	27 867.0	27 867.16	-0.16	c
$3_{1,2} \leftarrow 2_{1,1}$	29 522.8	29 522.71	0.09	c
$3_{2,2} \leftarrow 2_{2,1}$	28 699.3	28 699.35	-0.05	c
$3_{1,2} \leftarrow 3_{0,3}$	38 814.0	38 814.00	0.00	c
$4_{0,4} \leftarrow 3_{0,3}$	38 203.0	38 203.11	-0.11	c
$4_{1,4} \leftarrow 3_{1,3}$	37 149.0	37 148.97	0.03	c
$4_{1,3} \leftarrow 3_{1,2}$	39 356.2	39 356.09	0.11	c
$4_{2,3} \leftarrow 3_{2,2}$	38 260.9	38 260.81	0.09	c
$4_{2,2} \leftarrow 3_{2,1}$	38 322.3	38 322.27	0.03	c
$4_{3,2} \leftarrow 3_{3,1}$	38 279.1	38 279.04	0.06	c
$4_{3,1} \leftarrow 3_{3,0}$		38 279.29	-0.19	c
$4_{1,3} \leftarrow 4_{0,4}$	39 966.8	39 966.98	-0.18	c
$5_{1,4} \leftarrow 5_{0,5}$	41 442.2	41 442.27	-0.07	c
$6_{0,6} \leftarrow 5_{1,5}$	24 015.85	24 015.81	0.04	d
$7_{1,7} \leftarrow 6_{0,6}$	96 629.70	96 629.70	0.00	d
$7_{0,7} \leftarrow 6_{1,6}$	34 942.09	34 942.24	-0.15	d
$8_{2,6} \leftarrow 8_{1,7}$	102 871.77	102 871.77	0.00	d
$7_{2,6} \leftarrow 8_{1,7}$	25 137.23	25 137.32	-0.09	d
$8_{2,6} \leftarrow 9_{1,9}$	39 275.26	39 275.25	0.01	d

TABLE I—*Continued*

Transition	Observed Frequency	Calculated Frequency[a]	Deviation[b]	Ref.
$10_{2,8} \leftarrow 9_{2,7}$	96 486.87	96,486.87	0.00	d,e
$10_{3,8} \leftarrow 9_{3,7}$	95 782.68	95 782.63	0.05	d
$10_{3,7} \leftarrow 9_{3,6}$	95 816.46	95 816.38	0.08	d
$10_{4,7} \leftarrow 9_{4,6}$	95 743.95	95 743.69	0.26	d
$10_{4,6} \leftarrow 9_{4,5}$		95 744.12	-0.17	d
$10_{5,6} \leftarrow 9_{5,5}$	95 724.00	95 723.98	0.02	d
$10_{5,5} \leftarrow 9_{5,4}$				
$10_{6,5} \leftarrow 9_{6,4}$	95 717.67	95 717.55	0.12	d
$10_{6,4} \leftarrow 9_{6,3}$				
$10_{7,4} \leftarrow 9_{7,3}$	95 717.67	95 717.87	-0.20	d
$10_{7,3} \leftarrow 9_{7,2}$				
$10_{8,2} \leftarrow 9_{8,1}$	95 722.17	95 722.31	-0.14	d
$10_{8,3} \leftarrow 9_{8,2}$				
$10_{2,8} \leftarrow 10_{1,9}$	99 527.22	99 527.26	-0.04	d
$9_{2,7} \leftarrow 10_{1,10}$	33 312.64	33 312.71	-0.07	d
$11_{0,11} \leftarrow 10_{0,10}$	103 922.97	103 923.03	-0.06	d,e
$11_{1,11} \leftarrow 10_{1,10}$	101 883.26	101 883.26	0.00	d,e
$11_{1,10} \leftarrow 10_{1,9}$	107 895.51	107 895.65	-0.14	d,e
$11_{2,10} \leftarrow 10_{2,9}$	105 011.71	105 011.86	-0.15	d
$11_{2,9} \leftarrow 10_{2,8}$	106 306.77	106 306.80	-0.03	d
$11_{3,9} \leftarrow 10_{3,8}$	105 380.13	105 380.19	-0.06	d
$11_{3,8} \leftarrow 10_{3,7}$	105 434.94	105 434.91	0.03	d
$11_{5,7} \leftarrow 10_{5,6}$	105 304.53	105 304.50	0.03	d
$11_{5,6} \leftarrow 10_{5,5}$				
$11_{6,6} \leftarrow 10_{6,5}$	105 294.36	105 294.33	0.03	d
$11_{6,5} \leftarrow 10_{6,4}$				

TABLE I—*Continued*

Transition	Observed Frequency	Calculated Frequency[a]	Deviation[b]	Ref.
$11_{7,5} \leftarrow 10_{7,4}$ $11_{7,4} \leftarrow 10_{7,3}$	105 292.83	105 292.86	-0.03	d
$11_{8,4} \leftarrow 10_{8,3}$ $11_{8,3} \leftarrow 10_{8,2}$	105 296.46	105 296.57	-0.11	d
$11_{2,9} \leftarrow 11_{1,10}$	97 938.33	97 938.41	-0.08	d
$10_{2,8} \leftarrow 11_{1,11}$	27 916.31	27 916.32	-0.01	d
$12_{2,10} \leftarrow 12_{1,11}$	96 484.74	96 484.72	0.02	d
$11_{2,9} \leftarrow 12_{1,12}$	23 137.86	23 137.89	-0.03	d
$12_{1,11} \leftarrow 11_{2,10}$	23 963.56	23 963.52	0.04	d
$13_{1,13} \leftarrow 12_{0,12}$	140 462.40	140 462.25	0.15	d
$13_{0,13} \leftarrow 12_{1,12}$	102 091.14	102 091.02	0.12	d
$15_{0,15} \leftarrow 14_{0,14}$	140 456.20	140 456.11	0.09	d
$14_{3,12} \leftarrow 15_{2,13}$	32 150.12	32 150.13	-0.01	d
$16_{3,13} \leftarrow 17_{2,16}$	29 859.90	29 859.84	0.06	d
$17_{3,14} \leftarrow 18_{2,17}$	22 155.03	22 155.07	-0.04	d
$20_{2,18} \leftarrow 20_{1,19}$	96 404.01	96 403.90	0.11	d
$20_{2,18} \leftarrow 19_{3,17}$	31 944.58	31 944.64	-0.06	d
$21_{2,19} \leftarrow 21_{1,20}$	98 528.76	98 528.61	0.15	d
$22_{2,20} \leftarrow 22_{1,21}$	101 246.22	101 246.10	0.12	d
$22_{4,18} \leftarrow 23_{3,21}$	38 984.32	38 984.36	0.04	d
$22_{4,19} \leftarrow 23_{3,20}$	29 626.54	29 626.59	-0.05	d
$23_{2,21} \leftarrow 24_{1,24}$	27 169.25	27 169.22	0.03	d
$23_{4,19} \leftarrow 24_{3,22}$	29 735.40	29 735.39	0.01	d
$26_{2,25} \leftarrow 25_{3,22}$	21 869.23	21 869.15	0.08	d
$27_{2,26} \leftarrow 26_{3,23}$	24 364.33	24 364.33	0.00	d

TABLE I—*Continued*

Transition	Observed Frequency	Calculated Frequency[a]	Deviation[b]	Ref.
$28_{2,27} \leftarrow 27_{3,24}$	26 025.07	26 024.99	0.08	d
$28_{3,25} \leftarrow 27_{4,24}$	32 998.37	32 998.45	-0.08	d
$29_{2,28} \leftarrow 28_{3,25}$	26 814.38	26 814.40	-0.02	d
$30_{2,29} \leftarrow 29_{3,26}$	26 703.69	26 703.75	-0.06	d

[a] Calculated frequencies were obtained using more significant figures than the rotational parameters of Table II.

[b] Deviation = observed frequency - calculated frequency.

[c] D. J. Meschi and R. J. Myers, ref. (1).

[d] Present work.

[e] Observed as discharge product of SO_2 and S_2.

$b_p = (\mathcal{C} - \mathcal{B})/(2\mathcal{A} - \mathcal{B} - \mathcal{C})$ is Wang's asymmetry parameter appropriate for a near prolate top. The rotational constants, $\mathcal{A}$, $\mathcal{B}$, and $\mathcal{C}$, differ from the usual effective rotational constants A, B, and C, by an additional centrifugal distortion contribution which depends on the distortion coefficient $R_6 = -2.606 \times 10^{-5}$ MHz. For S_2O this contribution is very small.

The distortion constants given in Table II were obtained from a least-squares analysis of the 70 rotational transitions listed in Table I. The details of the analysis have been outlined previously by Cook, DeLucia, and Helminger (*11*). All calculations were made with 16 digit floating point arithmetic. The experimental error in the measured frequencies is considered to be around 0.1 MHz. The distortion effects are small for the transitions observed, and no P^6 dependence was required to fit the spectrum. The uncertainties in the constants listed in Table II are twice the statistical standard deviations. The standard deviation of the least-squares fit is 0.10 MHz. The largest correlation coefficient between the distortion constants, $\rho = 0.71$, occurred for the constants Δ_J and Δ_{JK} (*12*).

An analysis which omitted the millimeter wave transitions was also carried out. The uncertainties in the constants and the correlation coefficients were significantly higher for this analysis than for the analysis of the complete data set. Nevertheless, the distortion constants obtained were within the final quoted statistical errors, except for Δ_J and Δ_{JK} which were outside these limits but within three times the quoted uncertainties.

TABLE II

ROTATIONAL AND DISTORTION CONSTANTS OF S_2O (MHz)[a]

$\mathcal{A} = 41915.44 \pm 0.03$	$A = 41915.44^{b}$
$\mathcal{B} = 5059.101 \pm 0.003$	$B = 5059.07^{b}$
$\mathcal{C} = 4507.163 \pm 0.003$	$C = 4507.19^{b}$
$\Delta_J = 1.895 \times 10^{-3} \pm 0.005 \times 10^{-3}$	$\Delta_{JK} = -3.192 \times 10^{-2} \pm 0.008 \times 10^{-2}$
$\Delta_K = 1.197 \times 10^{0} \pm 0.002 \times 10^{0}$	$\delta_J = 3.453 \times 10^{-4} \pm 0.008 \times 10^{-4}$
$\delta_K = 1.223 \times 10^{-2} \pm 0.012 \times 10^{-2}$	

[a] The errors quoted are for 95% confidence.

[b] Corrected for the effects of $R_6 = -(4\Delta_J + \tau_{bbcc})/32$, where τ_{bbcc} was calculated from the planar relations.

The distortion coefficients of Table II are linear combinations of the τ-distortion-constants of Kivelson and Wilson *(13)*. To relate the distortion constants to the molecular force field, it is convenient to switch from the distortion constants of Table II to the τ-distortion constants. The three τ's, viz., τ_{aaaa}, τ_{bbbb}, and τ_{cccc}, and Watson's *(10)* two linear combinations of τ's,

$$\tau_1 = \tau_{bbcc} + \tau_{aacc} + \tau'_{aabb}, \tag{2}$$

$$\tau_2 = A\tau_{bbcc} + B\tau_{aacc} + C\tau'_{aabb}, \tag{3}$$

with $\tau'_{aabb} = \tau_{aabb} + 2\tau_{abab}$, may be evaluated directly from the constants of Table II *(11)*. These are given in Table III. To evaluate the distortion constants, τ_{aabb}, τ_{aacc}, τ_{bbcc}, or τ_{abab}, the planar relations must be employed [see, e.g., *(11)*]. Because of these relations for a planar molecule, there are only four independent distortion constants, and for discussion of the force field we chose the constants τ_{aaaa}, τ_{bbbb}, τ_{cccc}, and τ_{abab}. Since five distortion coefficients have been evaluated from the spectrum, there are various ways of calculating τ_{abab} from the data. Because of vibrational effects the results of the various calculations will not be quite the same. The constants obtained by three different procedures of calculation are given in Table III. In one procedure τ_{aabb}, τ_{bbcc}, and τ_{aacc} are calculated via the planar relations from the constants, τ_{aaaa}, τ_{bbbb}, and τ_{cccc}. These are then used in Eq. (2) to evaluate τ_{abab} from τ_1. This gives the first entry

TABLE III

ALTERNATE QUARTIC DISTORTION CONSTANTS[a]

$\tau_{aaaa} = -4.669 \pm 0.008$	$\tau_{bbbb} = -0.01034 \pm 0.00002$
$\tau_{cccc} = -0.00482 \pm 0.00002$	$\tau_1 = 0.1049 \pm 0.0003$
$\tau_2 = 248.2 \pm 1.7$	$\tau_{abab} = \begin{cases} -0.0307^{b,c} \\ -0.0304^{b,d} \\ -0.0324^{b,e} \end{cases}$

[a] All entries in units of MHz except τ_2 which has units of $(\text{MHz})^2$.

[b] Calculated by means of the planar relations. See text.

[c] From τ_1.

[d] From τ_2.

[e] From (τ_1, τ_2).

TABLE IV

VIBRATIONAL ASSIGNMENT OF S_2O

Vibrational frequencies[a] (cm^{-1})		Force constants (mdyn/Å)	
ω_1	1165	F_{11}	8.34
ω_2	679	F_{22}	4.27
ω_3	387	F_{33}	0.55

	Distortion constants (MHz) Observed	Calculated
τ_{aaaa}	-4.669	-4.119
τ_{bbbb}	-0.0103	-0.0109
τ_{cccc}	-0.0048	-0.0053
τ_{abab}	-0.03	-0.029

[a] ω_1: S–O, ω_2: S–S, ω_3: $\sphericalangle$ SSO.

in Table III. A similar calculation can be made using τ_2 rather than τ_1. In an alternate procedure, τ_{bbcc} is calculated from the planar relations and employing this constant τ_1 and τ_2 are solved simultaneously for τ'_{aabb} and τ_{aacc}. The τ_{abab} can then be extracted from τ'_{aabb} once τ_{aabb} is calculated from the planar relations. It should be pointed out that in the planar relations and subsequent calculations, the effective rotational constants have been employed.

For the equilibrium parameters, the following relation given by Watson (*10*) holds for a planar molecule:

$$\tau_2 = C\tau_1 + (A + B)\tau_{cccc}. \tag{4}$$

The value of τ_2 calculated from this expression is 246.6 ± 1.8 $(\text{MHz})^2$, while the observed value is 248.2 ± 1.7 $(\text{MHz})^2$. Apparently, for S_2O this planarity-constraint equation is reasonably well satisfied.

IV. VIBRATIONAL ASSIGNMENT

The infrared spectrum of the discharge products of a mixture of sulfur and SO_2 has been investigated by Jones (*4*). Two absorption bands were observed which he believed arose from the species S_2O_2. With the work of Meschi and Myers (*1*), it is reasonable to assign the infrared absorption bands at 1165 and 679 cm^{-1} to the S–O and S–S stretching modes of S_2O. The infrared spectrum of the bending mode does not seem to have been published; however, Meschi and Myers (*1*) report in a footnote that the bending mode has been observed by Blukis at 387 cm^{-1}. As far as we are aware, no further information on the infrared spectrum is available. It is, hence, interesting to see if these vibrational frequencies are consistent with the distortion data of disulfur monoxide. From the three vibrational frequencies the diagonal force constants may be evaluated and these are listed in Table IV. In reality, six sets of force constants may be evaluated from the observed vibrational frequencies and the results are similar to those obtained for the molecule NSF (*14*). The values given in Table IV are for the most reasonable assignment of 1165 cm^{-1} to the S–O stretching motion (S_1), 679 cm^{-1} to the S–S stretching motion (S_2), and 387 cm^{-1} to the bending motion (S_3). The τ's are directly related to elements of the inverse force constant matrix and may, hence, be calculated from the force constants of Table IV. In particular,

$$\tau_{\alpha\beta\gamma\delta} = -\frac{(h/8\pi^2)^2}{RI_{\alpha\alpha}I_{\beta\beta}I_{\gamma\gamma}I_{\delta\delta}} \sum_{i,j} J_{\alpha\beta}{}^{(i)}(F^{-1})_{ij}J_{\gamma\delta}{}^{(j)}, \qquad (5)$$

with $R = 10^{-22}/2h$, $J_{\alpha\beta}{}^{(i)} = (\partial I_{\alpha\beta}/\partial S_i)$. The $J_{\alpha\beta}{}^{(i)}$ depend on the masses and geometry of the molecule. Employing atomic mass units, bond distances in angstroms, $(h/8\pi^2 I_{\alpha\alpha})$ in units of MHz, and bond stretching force constants in dyn/cm, bond bending force constants in dyn-cm, etc., the τ's are given in MHz units. The masses and fundamental constants have been taken from (*15*) and the molecular geometry: r(S–O) = 1.465 Å, r(S–S) = 1.884 Å, and ∠(SSO) = 118° from (*1*). The calculated values of the distortion constants are compared with the observed values in Table IV. The agreement is good and lends support to the vibrational assignment discussed previously. Although one of the interaction constants could possibly be obtained by combining the infrared and microwave data, it does not seem warranted at this time.

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