NOTE

High-Resolution Fourier Transform Infrared Spectroscopy of the NS Radical

High-resolution Fourier transform spectroscopy (FTS) has recently been employed in our laboratory for obtaining infrared absorption spectra of several chemically short-lived species (l-5). The precision of the line positions obtained via FTS is comparable to that of other high-resolution infrared techniques such as diode laser, difference frequency laser, and laser magnetic resonance spectroscopy. The FTS technique however, has the advantage of being able to record an entire spectrum simultaneously, thus allowing a consistent set of line positions as well as intensities to be recorded.

In the present work we report high-resolution infrared FTS absorption measurements of the NS $(X^2\Pi)$ radical. The NS radical has been previously studied by several groups using diode laser, microwave, millimeterwave, and UV spectroscopy (6-12). The present study extends the measurements for v=0 and v=1 of the $X^2\Pi$ state to higher J, thus allowing a more precise determination of the molecular constants.

Details of the apparatus used in this study have been presented earlier (I-5, I3). The NS radical was made in three separate sidearms of the absorption cell. The sidearms were spaced 55 cm apart along the length of the absorption cell with the first sidearm located 10 cm in front of the first multipass mirror. The multipass configuration provided an effective absorption path length of ~ 100 m. The NS radical was generated by passing a N₂/SCl₂ mixture through a microwave discharge (6). The optimum absorption signal was obtained using maximum available pumping speed, with a cell residence time of ~ 0.08 sec, and at a total pressure of 0.5 Torr. The absorption spectrum, shown in Fig. 1, contains transitions up to J = 32.5 for the ${}^{2}\Pi_{3/2}$ spin-orbit state and J = 35.5 in the ${}^{2}\Pi_{1/2}$ spin-orbit state. The observed transitions are listed in Table

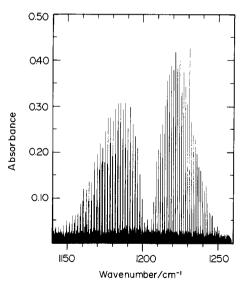


Fig. 1. High-resolution Fourier transform infrared absorption spectrum of the NS radical. The spectrum was recorded at 0.005 cm⁻¹ resolution in 61 coadded scans.

TABLE I $\label{eq:TABLE I}$ Observed Transitions for the v = 1–0 Band of $^{14}N^{32}S$ (cm $^{-1})$

a) ² J	R(J)		o-c ^a	P(J)	0-C			
								
1.5	1207.920		-47					
2.5	1209.42		10	1200.201				
3.5	1210.91		-4	1198.618				
4.5	1212.40		28	1197.025				
5.5	1213.866		-52 - 8	1195.417				
5.5 7.5	1215.321 1216.76		26	1193.798 1192.166				
3.5	1218.19		13	1190.52				
9.5	1219.60		7	1188.865				
0.5	1221.00		32	1187.195				
1.5	1222.39		-14	1185.513				
2.5	1223.77	508	-34	1183.819	980 -36			
3.5	1225.13	869	0	1182.113	348 -44			
4.5	1226.48	882	2	1180.395	550 10			
5.5	1227.82	617	43	1178.664				
6.5		-	-	1176.921				
7.5		-	-	1175.166				
8.5		-	-	1173.399				
9.5 0.5	1234.31	204	47	1171.619				
1.5	1235.56		-41	1169.824				
2.5	1236.81		7	1166.209				
3.5	1238,04		-27	1164.38				
4.5	1239.25		-28	1162.542				
5.5	1240.46		-13	1160.690				
6.5	1241.65	274	- 7	1158.827				
7.5	1242.829		10	1156.951	L 8 5 - 32			
8.5	1243.99	209	44	1155.065	519 11			
9.5	1245.140		-31					
0.5 1.5	1246.27 1247.39		-25 -12					

ь) ²	Transit	ions					***************************************	
ь) ²	Transit	ions O-C	R _f (3)	o-c	P _e (J)	D-C	$P_{f}^{(J)}$	0-
.1 0.5	R _e (J)	0-C -2	R _f (J)	0-C 24	P _e (J)	O-C	P _f (J)	0-
.1 0.5 1.5	R _e (J) 1206.55190 1208.05998	0-C -2 16	1206.56540 1208.07315	24 8	1201.96539	O-C	P _f (J)	0-
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a Observed minus Calculated x10⁵.

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TABLE II						
Molecular Constants for ¹⁴ N ³² S (cm ⁻¹)						

Constant	v = 0	v = 1		
A	[223.15]	222,96490(6) ^a		
^ _D ×10 ⁻³	0.1180(12)	0.1085(12)		
В	0.7723812(13)	0.7660851(13)		
D x10 ⁻⁵	0.12577(10)	0.126156(90)		
P	0.013197(70)	0.013215(70)		
p _D x10 ⁻⁷ q x10 ⁻⁶	-0.86(43)	[-0.86]		
q x10 ⁻⁶	[0.10]	[0.22]		
٥ م		1204.18222(4)		

Values in brackets were fixed in the fit (see text).

I. Absolute frequency calibration was obtained using the infrared spectrum of N_2Ov_3 band as a reference (14).

The observed line positions were fit using the Hamiltonian given by Zare et al. (15). This Hamiltonian, when specialized to ²II systems, takes the form

$$\langle {}^{2}\Pi_{3/2}|H|{}^{2}\Pi_{3/2}\rangle = \frac{1}{2}A + \frac{1}{2}q - D + (B - D + \frac{1}{2}q + \frac{1}{2}A_{D})(Z^{2} - 2) - D(Z^{2} - 2)^{2}$$

$$\langle {}^{2}\Pi_{1/2}|H|{}^{2}\Pi_{1/2}\rangle = -\frac{1}{2}A + D + (B - D - \frac{1}{2}A_{D})Z^{2} - DZ^{4}$$

$$+ \frac{1}{2}(p + \frac{1}{2}p_{D}(Z^{2} - \frac{1}{4}))(1 \pm Z) + \frac{1}{2}q(1 \pm Z)^{2} - \gamma$$

$$\langle {}^{2}\Pi_{3/2}|H|{}^{2}\Pi_{1/2}\rangle = -B - \frac{1}{4}p - \frac{1}{4}p_{D}(Z^{2} - \frac{1}{4}) - \frac{1}{2}q(1 \pm Z)$$

$$+ 2D(Z^{2} - 1)(Z^{2} - 1)^{1/2} - (\gamma/2)(Z^{2} - 1)^{1/2},$$
(3)

where $Z=J+\frac{1}{2}$, A is the spin-orbit interaction parameter, A_D is the centrifugal distortion correction to A, γ is the spin-rotation parameter, p and q are Λ -doubling parameters, p_D is the centrifugal distortion correction to p, and p and p are the rotational and centrifugal distortion parameters, respectively. The \pm signs distinguish between the Λ -doubling components of a given rotational level. When Λ doubling was not resolved, the observed line positions were fitted to the mean of the calculated Λ -doublet splitting. Brown and Watson (16) have shown that γ and A_D are highly correlated and cannot be determined simultaneously. Therefore, we have arbitrarily chosen to set $\gamma=0$ and allow A_D to vary in the fits.

As observed in previous studies (6, 7), Λ doubling could be resolved only for the ${}^2\Pi_{1/2}$ spin-orbit state. Consequently, the Λ -doubling parameters p, p_D , and q were not all simultaneously determinable from the fit. In order to circumvent this difficulty, the value of q was fixed to that given in Ref. (10). Preliminary fits showed that the change in the parameter P_D was not statistically significant in going from v=0 to v=1. Thus, $\Delta P_D = P_D (v=1) - P_D (v=0)$ was set equal to zero. Since we did not observe any of the extremely weak ${}^2\Pi_{3/2} \leftrightarrow {}^2\Pi_{1/2}$ transitions, the measurements did not allow for an accurate determination of the spin-orbit constant. As a result, the value of the spin-orbit constant A(v=0), was fixed to the results of the UV measurements of Ref. (12). With A(v=0) fixed, our analysis provides a determination of ΔA . All transitions included in the fitting were weighted equally.

The quality of the fit, as exhibited by the residuals (obs. – cal.), is shown in Table I and the molecular constants obtained from the fit are given in Table II. The rms standard deviation of the fit is 0.00026 cm⁻¹ and is almost a factor of five better than the diode laser study of Matsumura *et al.* (6). The molecular constants are in good agreement with previous measurements.

a Values in parentheses are one standard deviation.

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AMITABHA SINHA ¹
JAMES B. BURKHOLDER
PHILIP D. HAMMER ²
CARLETON J. HOWARD

NOAA Aeronomy Laboratory
R/E/AL2
325 Broadway
Boulder, Colorado 80303
and
Cooperative Institute for Research in Environmental Sciences
University of Colorado
Boulder, Colorado 80303
Received March 24, 1988

¹ Present address: Department of Chemistry, University of Wisconsin, Madison, WI 53706.

² Present address: NASA, Ames Research Center, Moffett Field, CA 94035.