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Quadrupole transitions of the $1 \leftarrow 0$ band of N_2 observed in a high resolution atmospheric spectrum

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Résumé. — Un spectre d'absorption à haute résolution de l'atmosphère terrestre enregistré à Kitt Peak à l'aide d'un spectromètre par transformation de Fourier a permis d'observer les transitions quadrupolaires S7 à S16 de la bande $1 \leftarrow 0$ de N_2 . L'analyse des largeurs équivalentes et des profondeurs centrales de ces raies conduit, d'une part à la détermination de la dérivée du moment quadrupolaire $(\partial Q/\partial r)_e = (0,94 \pm 0,05) ea_0$ et d'autre part, à une estimation du coefficient d'élargissement par l'air $\gamma_{N_2-air}^0 = 0,06 \pm 0,02 \text{ cm}^{-1} \text{ atm.}^{-1}$ à 296 K. En outre l'identification des raies telluriques observées entre $2\,391,5$ et $2\,467 \text{ cm}^{-1}$ est donnée.

Abstract. — Using a high resolution atmospheric absorption spectrum recorded at Kitt Peak with the solar Fourier transform spectrometer, it has been possible to observe the quadrupole transitions S7 to S16 of the $1 \leftarrow 0$ band of N_2 . The analysis of the equivalent widths and of the central depths of these lines has enabled us to determine the derivative of the quadrupole moment $(\partial Q/\partial r)_e = (0.94 \pm 0.05) ea_0$ and to estimate the broadening coefficient by air $\gamma_{N_2-air}^0 = 0.06 \pm 0.02 \text{ cm}^{-1} \text{ atm.}^{-1}$ at 296 K. Finally the assignment of telluric lines between $2\,391.5$ and $2\,467 \text{ cm}^{-1}$ is also given.

1. **Introduction.** — Fourier transform spectroscopy is able to produce spectra of very high resolution as well as very high signal to noise ratio. These qualities are very useful in atmospheric physics since it is now possible to detect very weak absorptions due to the most intense lines of trace gases or due to the weakest lines of more abundant constituents. The atmospheric spectrum between $2\,391.5$ and $2\,467 \text{ cm}^{-1}$ studied in this work has been recorded with the solar Fourier transform spectrometer of Kitt Peak National Observatory (2 078 m) with a resolution of 0.009 cm^{-1} using the sun as a source. This spectral region which lies just above the end of the very strong $CO_2 \nu_3$ band (see Fig. 1) is influenced by the CO_2 continuum and to a lesser extent by the N_2 continuum but is relatively free of any strong lines. These conditions are favourable to search for the quadrupole lines of the $1 \leftarrow 0$

band of N_2 the intensity of which is expected to be very small.

2. **Analysis.** — The spectrum was recorded around 11.00 AM (local time) on October 25, 1979 at an average secant of 1.475. The ground pressure was 798 mbars and the ground temperature $23^\circ C$. A portion of the spectrum is shown in figure 1. Using the AFGL line parameter compilation [1] the CO_2 , N_2O and CH_4 telluric lines were first identified but some strong enough lines were left unassigned. Among them, some lines much broader than the telluric ones (see Fig. 2) were tentatively attributed to the sun. Some others were without any doubt of telluric origin because their profile was the result of the superposition of a wide tropospheric contribution and of a narrow stratospheric one. Using the

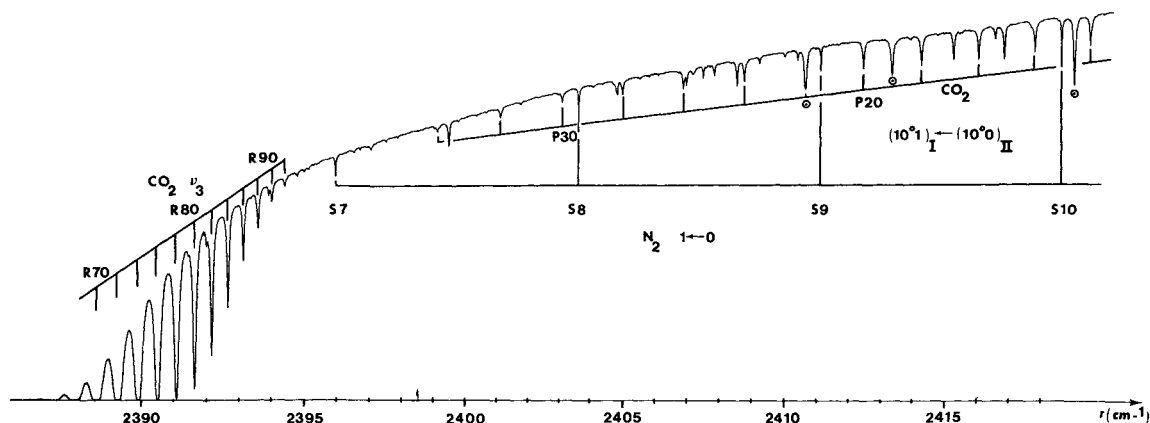


Fig. 1. — Portion of the atmospheric spectrum between 2387 and 2420 cm^{-1} . The lines S7 to S10 of the $1 \leftarrow 0$ quadrupole band of N_2 are easily seen superimposed on the CO_2 and N_2 continua. Some CO_2 and solar lines also appear.

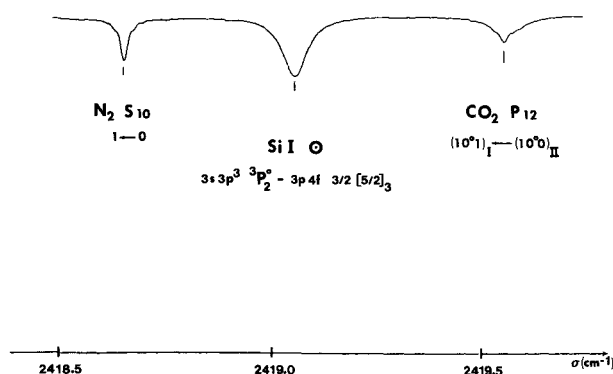


Fig. 2. — Detailed portion of the atmospheric spectrum showing the S10 quadrupole line of the $1 \leftarrow 0$ band of N_2 together with a Si I solar line and a CO_2 telluric line. Observe the very different line profiles: The solar line is very broad; when comparing the N_2 and CO_2 lines, the stratospheric part of the N_2 line is more prominent because the lower state energy of the CO_2 transition is much higher (hot band).

results obtained by Bendtsen [2] on the Raman spectrum of N_2 it has been possible to assign these lines to the quadrupole transitions S7 to S16 of the $1 \leftarrow 0$ band of the nitrogen molecule ⁽¹⁾. We have gathered in table I the list of the lines observed in the spectrum with their position, assignment and central depth.

3. **Results.** — It is important to deduce from our atmospheric spectrum quantitative results on the intensity of the N_2 lines because it seems difficult to obtain in the laboratory an optical density $P \times l$ comparable to what can be reached using the atmosphere as the absorbing medium

$$(P \times l \simeq 5 \times 10^5 \text{ atm. cm}).$$

Thus, we have performed a quantitative analysis based on a simplified atmospheric model (constant temperature gradient of 6 K/km in the troposphere,

⁽¹⁾ The S6 line is barely visible between the very strongly absorbing R68 and R70 ν_3 lines of CO_2 .

constant temperature of 209 K above 15 km in the stratosphere) to reproduce the observed equivalent widths W and central depths H of the N_2 lines. From these quantities (see columns 5 and 6 of table II) we have deduced the intensities k_σ^N (in $\text{cm}^{-1}/\text{molecule cm}^{-1}$) at 296 K of 7 lines of the quadrupole $1 \leftarrow 0$ band and we have estimated an average broadening coefficient $\gamma_{\text{N}_2-\text{air}}^0 = 0.06 \pm 0.02 \text{ cm}^{-1} \text{ atm.}^{-1}$ at 296 K. This calculation is based on the equations:

$$\tau(\sigma') = \int_0^\infty k_\sigma^N(T) N(z) \Phi(T, P, \sigma') dz$$

$$W = \int_{\text{on the line}} [1 - \exp(-\tau(\sigma'))] d\sigma'$$

$$H = \int_{\text{on the line}} f(\sigma - \sigma') [1 - \exp(-\tau(\sigma'))] d\sigma'$$

where

— $N(z)$ is the number density at altitude z (in molecule cm^{-3});

— $\Phi(T, P, \sigma')$ is the normalized line profile:

$$\int \Phi(T, P, \sigma') d\sigma' = 1;$$

— $f(\sigma - \sigma')$ is the normalized instrumental profile:

$$\int f(\sigma - \sigma') d\sigma' = 1.$$

The line profile Φ is a Voigt profile involving the Doppler half-width $\gamma_D(T)$ and the Lorentz half-width

$$\gamma_L(T, P) = \gamma^0(T_0) \frac{P}{P_0} \left(\frac{T_0}{T} \right)^{1/2}.$$

The intensity of a quadrupole line $v', J' \leftarrow v, J$ [3] is given by:

$$k_\sigma^N(T) = \frac{8 \pi^5}{15 hc} \sigma^3 e^{-E''/kT} g_J \frac{(2J+1)}{Z(T)} \times \\ \times |\langle J 0 20 | J' 0 \rangle|^2 |\langle v | Q | v' \rangle|^2$$

Table I. — Atmospheric spectrum between 2 392 and 2 465 cm⁻¹.

The meaning of the different columns is : σ : Wavenumber (in cm⁻¹) of the line. For solar lines, because of the Doppler shift, the actual wavenumber is $\sigma \times 1.000\ 001\ 941\ 5$. P : Percentage of absorption at the centre of the line. The notation b in this column means that the corresponding line is blended in a stronger one ; in this case the neighbouring lines are joined by a brace.

Assignment : For telluric CO₂ or N₂O lines, except when explicitly stated, the vibrational assignment is : for CO₂ (10⁰)_I - (10⁰)_{II} ; for N₂O (12⁰) - (00⁰) or (13¹0) - (01¹0) when the c, d notation appears.

Telluric CH₄ lines belong to the P branch of the 2 ν_4 band. ⊙ means a solar line,) or) means that the line lies respectively downward or upward of the quoted position of the blend.

σ (cm ⁻¹)	P (%)	Assignment	σ (cm ⁻¹)	P (%)	Assignment	σ (cm ⁻¹)	P (%)	Assignment
2391.6520	94	CO ₂ ν_3 R 80	2431.6772	1.2	CO ₂ R 2	2449.8000	(b	N ₂ O P 9 c
2392.0050	4.8		2431.7680	2.3	CH ₄	2449.8412	(2.7	N ₂ O P 9 d + CH ₄
2392.1740	75	CO ₂ ν_3 R 82	2431.9780	0.8	N ₂ O P 29 c	2450.1072	(45	N ₂ O P 14
2392.6740	51	CO ₂ ν_3 R 84	2432.524	19	⊙?+N ₂ O P 34 + CH ₄	2450.2112	(b	CO ₂ R 30
2393.1480	30	CO ₂ ν_3 R 86	2432.8980	0.8	N ₂ O P 28 c	2450.4484	1.3	
2393.5976	18	CO ₂ ν_3 R 88	2433.1760	4.3	CO ₂ R 4	2450.6604	1.0	N ₂ O P 8 c
2393.9208	3.4		2433.3840	(13	N ₂ O P 33	2450.8480	(7.7	CH ₄
2394.0196	8.3	CO ₂ ν_3 R 90	2433.4640	(b	N ₂ O P 28 d	2450.9680	(47	N ₂ O P 13
2394.4176	4.1	CO ₂ ν_3 R 92	2433.5876	8.8	N ₂ S 12	2451.0732	(6.3	CH ₄
2395.9652	8.3	N ₂ S 7	2433.8120	0.9	N ₂ O P 27 c	2451.3400	2.3	CO ₂ R 32
2397.0472	1.9	CO ₂ P 36	2434.2840	16	N ₂ O P 32	2451.5192	1.3	N ₂ O P 7 c
2399.0644	2.6	CO ₂ P 34	2434.6492	5.4	CO ₂ R 6	2451.5396	1.3	N ₂ O P 7 d
2399.4228	8.7	⊙	2434.75	0.8	N ₂ O P 26 c	2451.8268	44	N ₂ O P 12
2401.0532	3.0	CO ₂ P 32	2435.1804	17	N ₂ O P 31	2452.2816	8.0	CH ₄
2403.0200	3.6	CO ₂ P 30	2435.6360	1.0	N ₂ O P 25 c	2452.3800	(b	N ₂ O P 6 c, d
2403.5656	12	N ₂ S 8	2435.8420	0.7		2452.4376	(2.3	CO ₂ R 34
2404.7640	(b	CH ₄	2436.0760	24	N ₂ O P 30 + CO ₂ R 8)	2452.6840	43	N ₂ O P 11
2404.7896	(4.5	CH ₄	2436.5412	1.1	N ₂ O P 24 c	2452.8456	7.0	CH ₄
2404.9600	4.5	CO ₂ P 28	2436.9676	22.	N ₂ O P 29	2453.2280	1.3	N ₂ O P 5 c, d
2406.8756	5.9	CO ₂ P 26	2437.4484	(b	N ₂ O P 23 c	2453.3540	1.0	
2406.9720	3.0	CH ₄	2437.5128	(7.0	CO ₂ R 10	2453.5388	42	N ₂ O P 10 + CO ₂ R 36
2407.1720	1.3		2437.8588	28	N ₂ O P 28 + X	2454.0760	1.1	N ₂ O P 4 c, d
2407.4880	3.4	CH ₄	2438.3464	1.1	N ₂ O P 22 c	2454.3924	39	N ₂ O P 9
2407.8428	3.4	CH ₄	2438.7460	26	N ₂ O P 27	2454.9212	0.8	N ₂ O P 3 c, d
2408.5480	7.9	CH ₄	2438.9040	6.6	CO ₂ R 12	2455.2440	37	N ₂ O P 8
2408.7648	6.6	CO ₂ P 24	2439.1372	0.9		2455.5364	0.8	
2409.2272	2.2		2439.2444	1.1	N ₂ O P 21 c	2455.7004	2.1	N ₂ S 15
2410.0140	1.4	⊙ Si I	2439.6320	28	N ₂ O P 26	2456.0944	34	N ₂ O P 7
2410.4160	3.0	CH ₄	2440.1408	1.1	N ₂ O P 20 c	2456.3684	1.6	
2410.6320	(b	CO ₂ P 22	2440.2680	6.9	CO ₂ R 14	2456.5550	2.4	
2410.6560	(11	⊙ Si I+CH ₄)	2440.4216	0.7	N ₂ O P 20 d	2456.9428	31	N ₂ O P 6
2411.1280	6.7	N ₂ S 9	2440.5160	30	N ₂ O P 25	2457.0740	8.9	⊙ Si I
2412.4660	7.9	CO ₂ P 20	2440.9976	(4.6	N ₂ S 13	2457.4488	1.3	N ₂ O (13 ¹ 0)-(01 ¹ 0) Q
2413.3610	10	⊙ Si I	2441.0308	(b	N ₂ O P 19 c	2457.7888	29	N ₂ O P 5 + ⊙ Si I
2414.2776	8.3	CO ₂ P 18	2441.2840	(b	N ₂ O P 19 d	2458.6344	22	N ₂ O P 4
2415.2824	7.2	⊙ ?	2441.3972	(33	N ₂ O P 24	2458.7940	1.6	
2416.0628	8.3	CO ₂ P 16	2441.4868	6.8	CH ₄	2459.4776	18	N ₂ O P 3
2416.5760	3.3	⊙ ?	2441.5992	(11	CH ₄ + CO ₂ R 16	2459.5744	8.4	CH ₄
2416.8600	9.0	⊙ ?	2441.6464	(b	CH ₄	2459.7740	8.3	CH ₄
2417.8212	8.1	CO ₂ P 14	2441.9216	1.1	N ₂ O P 18 c	2460.1360	13	⊙ ?
2418.6520	13	N ₂ S 10	2442.1460	1.1	N ₂ O P 18 d	2460.3192	11	N ₂ O P 2
2419.0576	18	⊙ Si I	2442.2772	34	N ₂ O P 23	2461.1588	5.8	N ₂ O P 1
2419.5524	7.8	CO ₂ P 12	2442.8100	1.1	N ₂ O P 17 c	2462.8332	5.3	N ₂ O R 0
2421.2572	7.0	CO ₂ P 10	2442.9152	8.8	CO ₂ R 18	2462.9936	3.6	N ₂ S 16
2421.4640	1.4	N ₂ O P 46	2443.0048	1.1	N ₂ O P 17 d	2463.2360	(15	⊙?+ N ₂ O R 6 c
2422.3996	1.5	N ₂ O P 45	2443.1552	37	N ₂ O P 22	2463.2848	(b	N ₂ O R 6 d
2422.7372	1.0		2443.6920	1.0	N ₂ O P 16 c	2463.6680	11	N ₂ O R 1
2422.9356	5.6	CO ₂ P 8	2443.8664	1.0	N ₂ O P 16 d	2463.8472	11	⊙ ?
2423.2560	2.5	CH ₄	2444.0308	38	N ₂ O P 21	2464.1172	1.6	N ₂ O R 7 d
2423.3132	5.1	X ? + CH ₄ + N ₂ O P 44)	2444.1976	5.3	CO ₂ R 20	2464.5004	17	N ₂ O R 2
2424.2560	2.1	N ₂ O P 43	2444.5724	1.0	N ₂ O P 15 c	2464.6848	0.7	CH ₄
2424.5860	4.5	CO ₂ P 6	2444.7240	1.0	N ₂ O P 15 d	2464.8632	1.2	N ₂ O R 8 c
2424.8780	3.6	⊙ Si I	2444.9040	40	N ₂ O P 20	2464.9496	1.6	N ₂ O R 8 d + CH ₄
2425.1800	3.2	N ₂ O P 42	2445.4536	8.0	CO ₂ R 22+N ₂ O P 14 c	2465.3320	22	N ₂ O R 3
2426.1028	(b	N ₂ O P 41	2445.7760	42	N ₂ O P 19	2465.6744	(b	N ₂ O R 9 c
2426.1392	(7.1	N ₂ S 11	2445.9724	9.5	⊙ Si I	2465.7164	(3.5	CH ₄
2426.2080	1.2	CO ₂ P 4	2446.3252	1.4	N ₂ O P 13 c	2465.7768	(b	N ₂ O R 9 d
2427.0212	4.3	N ₂ O P 40	2446.4340	1.4	N ₂ O P 13 d	2466.1608	28	N ₂ O R 4
2427.3180	1.1		2446.6460	44	N ₂ O P 18 + CO ₂ R 24)	2466.4800	1.0	N ₂ O R 10 c
2427.8052	0.8	CO ₂ P 2	2447.1988	1.4	N ₂ O P 12 c	2466.6092	11	⊙ ? + R 10 d + CH ₄)
2427.9380	5.1	N ₂ O P 39	2447.2880	1.4	N ₂ O P 12 d	2466.9884	34	N ₂ O R 5
2428.8200	(13	CH ₄	2447.5140	43	N ₂ O P 17			
2428.8480	(b	N ₂ O P 38	2447.8876	4.8	CO ₂ R 26			
2429.1568	5.9	CH ₄	2448.0688	1.4	N ₂ O P 11 c			
2429.5552	5.7	CH ₄	2448.1404	1.4	N ₂ O P 11 d			
2429.7640	7.6	N ₂ O P 37	2448.3800	46	N ₂ O P 16+N ₂ S 14			
2429.9360	1.2		2448.9348	1.4	N ₂ O P 10 c + CH ₄)			
2430.6720	8.5	N ₂ O P 36	2448.9960	1.4	N ₂ O P 10 d			
2431.0680	9.2	CH ₄	2449.0856	9.1	⊙ Si I+CO ₂ R 28			
2431.5796	13	N ₂ O P 35 + CH ₄	2449.2444	45	N ₂ O P 15			

Table II. — N₂ quadrupole vibration-rotation lines (1-0 band).

Assignment	$\sigma_{\text{obs.}}$ (cm ⁻¹)	$\sigma_{\text{calc.}}$ (cm ⁻¹) ^(a)	E'' (cm ⁻¹) ^(a)	W (10 ⁻³ cm ⁻¹)	P (%)	$k_{\sigma}^N(\text{obs.})$ (10 ⁻²⁸ cm ⁻¹ /molecule ⁻² at 296 K)	$k_{\sigma}^N(\text{calc.})$ ^(b)	$\left(\frac{\partial Q}{\partial r}\right)_e/ea_0$ ^(b)
S7	2 395.965 2	.969 1	111.398	3.43	8.3	1.78	1.57	1.00
S8	2 403.565 6	.568 5	143.219	5.58	11.7	2.97	3.04	0.93
S9	2 411.128 0	.130 5	179.015	2.23	6.7	1.22	1.42	0.87
S10	2 418.652 0	.654 8	218.783	4.84	12.6	2.76	2.60	0.97
S11	2 426.139 2	.141 1	262.523				1.15	
S12	2 433.587 6	.589 3	310.233	3.24	8.8	2.03	1.99	0.95
S13	2 440.9 976	.998 9	361.912				0.84	
S14	^(b)	2 448.369 8	417.557				1.38	
S15	2 455.700 4	.701 6	477.166	0.682	2.1	0.49	0.56	0.88
S16	2 462.993 6	.994 1	540.738	1.30	3.6	0.98	0.88	1.00

0.94 ± 0.05

^(a) Calculated from J. Bendtsen [2].

^(b) See text.

where

$Z(T)$ is the total partition function (for N₂
 $Z(296) = 465$).

g_J is the nuclear spin statistical weight (3 or 6 for J odd or even).

$\langle j_1 m_1 j_2 m_2 | jm \rangle$ is a Clebsch-Gordan coefficient and we have :

$$|\langle J 0 20 | J' 0 \rangle|^2 = \begin{cases} \frac{3J(J-1)}{2(2J-1)(2J+1)} & \text{for O lines } (J' = J-2) \\ \frac{J(J+1)}{(2J-1)(2J+3)} & \text{for Q lines } (J' = J) \\ \frac{3(J+1)(J+2)}{2(2J+1)(2J+3)} & \text{for S lines } (J' = J+2) \end{cases}$$

If one writes

$$Q = Q_e + \left(\frac{\partial Q}{\partial r}\right)_e \Delta r + \dots$$

one has to first order

$$\langle v | Q | v+1 \rangle = \left(\frac{\partial Q}{\partial r}\right)_e r_e \left(\frac{B_e}{\omega_e}\right)^{1/2} \sqrt{v+1}.$$

Using the following spectroscopic data [4] :

$$r_e = 1.097\,685 \text{ \AA}, \quad B_e = 1.998\,24 \text{ cm}^{-1}, \\ \omega_e = 2\,358.57 \text{ cm}^{-1}$$

for each line, we have determined from the observed intensity $k_{\sigma}^N(\text{obs.})$ a value of $(\partial Q/\partial r)_e/ea_0$ which is given in the last column of table II. The average value with its statistical errors is :

$$\left(\frac{\partial Q}{\partial r}\right)_e = (0.94 \pm 0.05) ea_0$$

which is in good agreement with the values 0.95 ea_0 [5] and 0.97 ea_0 [6] deduced from collision-induced fundamental transitions. Finally using our value of $(\partial Q/\partial r)_e$ we have computed the intensities $k_{\sigma}^N(\text{calc.})$ quoted in table II. In addition to its atmospheric interest, this work shows that, when measuring the continuum in the laboratory, it is necessary to take into account the contribution of the quadrupole lines which have not been observed in such experiments because of a higher pressure and a lower resolution.

Note. — After this manuscript was ready, we have been aware of a paper by Goldman *et al.* [A. Goldman, J. Reid and L. S. Rothman, *Geophys. Res. Lett.* **8** (1981) 77] reporting the observation of O₂ quadrupole lines in the 1 600 cm⁻¹ region and mentioning the existence of N₂ quadrupole lines in the 2 400 cm⁻¹ region.

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