

Heterodyne Frequency Measurements on N₂O near 1060 cm⁻¹

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The results of heterodyne frequency measurements are given for the 10⁰⁰-02⁰⁰ band of N₂O centered at 1056 cm⁻¹. Nine lines are measured and fit with an rms deviation of 1.3 MHz. The data are combined with other infrared and microwave data in a least-squares fit that gives accurate ro-vibrational constants for the two states involved in these transitions. The analysis of the data is based on a treatment that includes the effect of *l*-type resonance between the 02⁰⁰ and 02²⁰ states. Derived tables of wavenumbers are given for the 02⁰⁰-00⁰⁰ band near 1168 cm⁻¹ and the 02⁰¹-00⁰⁰ band near 2460 cm⁻¹. © 1987 Academic Press, Inc.

INTRODUCTION

In order to provide accurate frequency calibration data using infrared transitions of N₂O (nitrous oxide), we have been making heterodyne frequency measurements on N₂O (*I-4*). In this paper we report frequency measurements on the very weak N₂O band centered near 1060 cm⁻¹ and due to the 10⁰⁰-02⁰⁰ transition.¹ This is the band that is in weak Fermi resonance with the 10⁰⁰-00⁰¹ band at 940 cm⁻¹ on which the N₂O laser is based.

We are not aware of any modern measurements of this band, but there are several good measurements of the 10⁰⁰-00⁰⁰ and 02⁰⁰-00⁰⁰ bands from which one can calculate the 10⁰⁰-02⁰⁰ band. Guelachvili (6) has given very good Fourier transform spectroscopy (FTS) measurements of the 02⁰⁰-00⁰⁰ band at 1168 cm⁻¹, but more recently Toth (7) has given more complete and (we believe) more accurate FTS measurements on the same band system. Aside from earlier measurements, the best direct measurements of the 10⁰⁰-00⁰⁰ transition near 2223 cm⁻¹ were made by Amiot and Guelachvili (8). The most accurate measurements involving the 10⁰⁰ state are the heterodyne frequency measurements of the 10⁰⁰-00⁰¹ transitions, which are the laser transitions of N₂O, by Whitford *et al.* (9). When combined with the heterodyne measurements on the 00⁰¹-00⁰⁰ transitions reported by Wells *et al.* (3), the 10⁰⁰ level is accurately located with respect to the ground state and based on frequency measurements alone.

¹ The vibrational numbering system adopted by the IAU-IUPAP joint commission on spectroscopy (5) is used throughout this paper. Most other authors use a notation that interchanges ν_1 and ν_3 .

We have been measuring systematically the frequencies of many transitions of N₂O in an effort to determine the energy separation of all the lower states of N₂O in order to calculate a consistent set of transition frequencies that can be used for frequency calibration in the infrared. This work has emphasized the use of frequency measurement techniques as a means of avoiding systematic errors that so easily affect wavelength measuring techniques such as interferometric techniques. Until now, we have not been able to obtain frequency measurements linking the 02⁰⁰ state to any of the other states, with the exception of a single frequency measurement of the *P*(75) 02⁰⁰-00⁰⁰ transition (2). While the 10⁰⁰-02⁰⁰ band is not so useful for some direct calibration purposes because of its low intensity, the present measurements were undertaken as a means of making accurate frequency measurements that would locate the 02⁰⁰ state with respect to the other low-lying vibrational states of N₂O.

Since the 10⁰⁰-02⁰⁰ transitions nicely overlap the region of the CO₂ laser transitions, it was possible to use a tunable diode laser and heterodyne frequency measurement techniques developed earlier in the NBS-Boulder Labs (10) to measure the 10⁰⁰-02⁰⁰ transitions against a well-characterized CO₂ laser. This in turn has enabled us to calculate accurate wavenumbers of the much stronger and therefore more useful 02⁰⁰-00⁰⁰ transitions near 1168 cm⁻¹. Furthermore, these data, when combined with the heterodyne measurements on the hot band transitions 02⁰¹-02⁰⁰ given by Hinz *et al.* (4), allow us to calculate the wavenumbers of the 02⁰¹-00⁰⁰ transitions near 2460 cm⁻¹.

EXPERIMENTAL DETAILS

The measurements in this experiment were relatively simple compared to the work in Refs. (2-4), in which a CO laser transfer oscillator was required. In this work only one CO₂ laser at a time was required for a heterodyne measurement. Most measurements used the normal ¹²C¹⁶O₂ laser; a few measurements required a ¹³C¹⁶O₂ laser. These lasers were constructed by the late F. R. Petersen, who used the Freed-Javan technique for stabilization (11). The estimated fractional-frequency uncertainty for these CO₂ lasers is about 1×10^{-9} , which is negligible relative to the other measurement uncertainties.

The basic technique was to lock the tunable diode-laser (TDL) frequency to the N₂O feature of interest and then heterodyne the TDL output with a nearby CO₂ laser transition in a HgCdTe mixer. For practical purposes, the measurement uncertainty was taken as one-tenth of the TDL-CO₂-laser beat-note linewidth plus a contribution due to the TDL lock scheme. This contribution was the Doppler width of the N₂O transition divided by the signal-to-noise ratio of the first derivative signal used for the TDL lock. In instances where the TDL power versus frequency output was not flat, the lock technique was suitably compensated (4). The procedure and spectrometer (with one or two exceptions) have not changed from the detailed account given in Ref. (10). One exception is that a closed-cycle cooler was used instead of a helium dewar. The other is that we no longer used the ethyl ether cells for isolation to prevent feedback (we did, however, remove the monochromator slits prior to locking the TDL and making the heterodyne frequency measurement). The particular TDLs used in

these measurements had comparatively slow tuning rates, and the laser linewidths were such as to permit 2- or 3-MHz uncertainties for most of the measurements indicated in Table I.

Since the transitions of interest were fairly weak, it was necessary to use pressures of about 0.67 kPa (5 Torr) in a 1.7-m cell and to heat the cell to temperatures in the 65 to 90°C range for some of the higher- J transitions.

ANALYSIS OF THE MEASUREMENTS

Since the number of heterodyne measurements we are able to make is rather limited, it is important to make use of other measurements that help to define the ro-vibrational constants for both states involved in these measurements. For the upper state (the 10^0_0 state) we have used the same data that were used in Ref. (3), namely, the heterodyne measurements of the laser transitions by Whitford *et al.* (9), the microwave measurements of Bogey (12), and appropriate combination differences taken from the FTS measurements of Amiot and Guelachvili (8).

In order to better define the constants for the lower state (the 02^0_0 state), the present measurements were combined with other infrared and microwave measurements involving the same state. Because there is a rather strong l -type resonance between the 02^0_0 and 02^2_0 states, measurements involving the latter state were also included in the fit. The microwave measurements compiled by Lovas and Tiemann (13) were used in the fit. We also used the heterodyne measurements given in Ref. (4) and the various infrared measurements mentioned therein, namely, the measurements by Guelachvili (6), Toth (7), Jolma *et al.* (14), and Kauppinen (15). These data were all combined in a nonlinear least-squares fit in which each value was weighted by the

TABLE I

Heterodyne Frequency Measurements on the 10^0_0 - 02^0_0 Band of Nitrous Oxide

CO ₂ Laser		Diff. Freq.	Nitrous Oxide		Obs.-Calc.
Isotope	Trans.	MHz	Trans.	Frequency, MHz ^a	MHz
¹³ C ¹⁶ O ₂	RII(28)	648.2	P(20)	31 094 136.7(40)	1.1
¹² C ¹⁶ O ₂	PII(24)	-2554.4	P(14)	31 270 692.8(40)	-0.5
¹² C ¹⁶ O ₂	PII(22)	-1440.4	P(12)	31 327 521.1(40)	-0.2
¹² C ¹⁶ O ₂	PII(16)	+403.1	P(6)	31 491 840.5(80)	-1.0
¹² C ¹⁶ O ₂	PII(14)	+512.8	P(4)	31 544 541.7(40)	-2.3
¹² C ¹⁶ O ₂	RII(10)	+549.9	R(21)	32 134 816.8(40)	0.0
¹² C ¹⁶ O ₂	RII(18)	+2126.0	R(30)	32 298 843.1(40)	1.1
¹² C ¹⁶ O ₂	RII(24)	+2582.6	R(37)	32 412 768.3(40)	0.1
¹² C ¹⁶ O ₂	RII(30)	-1317.7	R(44)	32 515 225.9(70)	2.7

^a The uncertainty in the last digits is given in parentheses.

TABLE II

N₂O Rotational Constants^a (in cm⁻¹) Used in the Present Analysis

Vib. State	B	Dx10 ⁷	Hx10 ¹³
00 ⁰ ₀	0.419010993(16)	1.760727(170)	-0.1787(324)
10 ⁰ ₀	0.415559533(19)	1.754940(275)	-0.084(73)
02 ⁰ ₀	0.419919782(24)	1.869710(406)	-1.547(189)
02 ² ₀ ^b	0.420124825(19)	1.817151(364)	1.014(253)
02 ⁰ ₁	0.41814807(51)	1.89653(242)	----
02 ² ₁ ^c	0.41852996(80)	1.75419(793)	2.29(217)
	qx10 ⁴	q _J x10 ⁹	q _{JJ} x10 ¹⁴
020	7.60818(90)	2.8157(1033)	-4.85(167)
021	8.2508(282)	3.436(1154)	----

a) The uncertainty in the last digits (twice the standard error) is given in parentheses.

b) For the 02²₀ state $L = -1.99(40) \times 10^{-17}$ was also determined and the value $G_{\Delta} - G_{\Sigma} = 9.61215$ cm⁻¹.

c) The value of $G_{\Delta} - G_{\Sigma} = 12.80221$ cm⁻¹ was used for the calculation of the l-type resonance between the 02⁰₁ and 02²₁ levels.

inverse square of its estimated uncertainty. In these fits the band center for each set of measurements was assumed to be different in order to uncouple systematic calibration errors in the different sets of measurements.

The 10⁰₀ energy levels, which are the upper state of the present measurements, were calculated in the least-squares fit by the equation

$$E = G_v + B_v J(J+1) - D_v [J(J+1) - l^2]^2 + H_v [J(J+1) - l^2]^3. \quad (1)$$

In the analysis of the lower state (02⁰₀) we used the following equations (16) in order to include the effect of the l-type resonance with the 02²₀ state,

$$E(02^0_0) = E_{\Sigma}^0 + \frac{1}{2}\delta - \frac{1}{2}\{\delta^2 + 4q^2[J^2(J+1)^2 - 2J(J+1)]\}^{1/2}, \quad (2)$$

$$E(02^2_0) = E_{\Delta}^0, \quad (3)$$

and

$$E(02^{2e}_0) = E_{\Delta}^0 - \frac{1}{2}\delta + \frac{1}{2}\{\delta^2 + 4q^2[J^2(J+1)^2 - 2J(J+1)]\}^{1/2}, \quad (4)$$

TABLE III

Band Centers Determined for N₂O

Vib. Transitions	ν_0 (cm ⁻¹)
10 ⁰ ₀ -02 ⁰ ₀	1055.624325(68)
02 ⁰ ₀ -00 ⁰ ₀	1168.132354(96)
02 ⁰ ₁ -02 ⁰ ₀	1293.863964(88)
02 ⁰ ₁ -00 ⁰ ₀	2461.996319(130)
10 ⁰ ₀ -00 ⁰ ₀	2223.756680(74)

a) The uncertainty in the last digits (twice the standard error) is given in parentheses.

where E_{Σ}^0 and E_{Δ}^0 are given by Eq. (1), with $l = 0$ and 2 , respectively,

$$\delta = E_{\Delta}^0 - E_{\Sigma}^0, \quad (5)$$

and

$$q = q_v - q_{vJ}J(J+1) + q_{vJJ}J^2(J+1)^2. \quad (6)$$

The observed transitions are then given by

$$\nu_{\text{obs}} = E' - E'' \quad (7)$$

and

$$\nu_o = G' - G''. \quad (8)$$

The various rotational constants determined by the fit and used later to calculate some transition wavenumbers are given in Table II. Table III gives the band centers determined either directly or indirectly by the present heterodyne frequency measurements.

TABLE IV

Wavenumbers (in cm^{-1}) Calculated^a for the 10^00-02^00 Band of N_2O

P-BRANCH	J''	R-BRANCH	P-BRANCH	J''	R-BRANCH
	0	1056.45544(3)	1021.18099(3)	35	1080.12759(3)
1054.78449(3)	1	1057.27784(3)	1020.05118(3)	36	1080.65456(3)
1053.93593(3)	2	1058.09150(3)	1018.91382(3)	37	1081.17367(3)
1053.07867(3)	3	1058.89644(3)	1017.76896(3)	38	1081.68496(3)
1052.21272(3)	4	1059.69265(3)	1016.61667(3)	39	1082.18850(4)
1051.33807(3)	5	1060.48014(3)	1015.45697(4)	40	1082.68430(4)
1050.45475(3)	6	1061.25891(3)	1014.28994(4)	41	1083.17242(4)
1049.56278(3)	7	1062.02896(3)	1013.11562(4)	42	1083.65291(4)
1048.66216(3)	8	1062.79031(3)	1011.93407(4)	43	1084.12580(4)
1047.75291(3)	9	1063.54296(3)	1010.74533(4)	44	1084.59114(4)
1046.83506(3)	10	1064.28693(3)	1009.54946(4)	45	1085.04898(4)
1045.90863(3)	11	1065.02222(3)	1008.34650(4)	46	1085.49934(4)
1044.97363(3)	12	1065.74885(3)	1007.13651(4)	47	1085.94228(4)
1044.03010(3)	13	1066.46684(3)	1005.91954(4)	48	1086.37784(4)
1043.07805(3)	14	1067.17621(3)	1004.69563(4)	49	1086.80605(4)
1042.11753(3)	15	1067.87697(3)	1003.46483(4)	50	1087.22696(5)
1041.14855(3)	16	1068.56915(3)	1002.22720(5)	51	1087.64061(5)
1040.17114(3)	17	1069.25277(3)	1000.98277(5)	52	1088.04702(5)
1039.18535(3)	18	1069.92784(3)	999.73159(5)	53	1088.44625(5)
1038.19120(3)	19	1070.59441(3)	998.47371(5)	54	1088.83832(6)
1037.18872(3)	20	1071.25249(3)	997.20917(6)	55	1089.22327(6)
1036.17797(3)	21	1071.90211(3)	995.93801(6)	56	1089.60113(6)
1035.15896(3)	22	1072.54330(3)	994.66028(6)	57	1089.97194(7)
1034.13175(3)	23	1073.17610(3)	993.37600(7)	58	1090.33573(7)
1033.09637(3)	24	1073.80053(3)	992.08523(7)	59	1090.69252(8)
1032.05287(3)	25	1074.41663(3)	990.78799(8)	60	1091.04236(9)
1031.00129(3)	26	1075.02444(3)	989.48433(8)	61	1091.38526(10)
1029.94166(3)	27	1075.62398(3)	988.17429(9)	62	1091.72125(11)
1028.87405(3)	28	1076.21531(3)	986.85788(10)	63	1092.05035(12)
1027.79849(3)	29	1076.79844(3)	985.53516(11)	64	1092.37261(13)
1026.71504(3)	30	1077.37343(3)	984.20614(12)	65	1092.68802(14)
1025.62373(3)	31	1077.94031(3)	982.87087(13)	66	1092.99663(16)
1024.52462(3)	32	1078.49913(3)	981.52937(15)	67	1093.29845(17)
1023.41776(3)	33	1079.04992(3)	980.18167(16)	68	1093.59350(19)
1022.30320(3)	34	1079.59272(3)	978.82780(18)	69	1093.88180(21)

a) The uncertainty in the last digits (one standard error) is given in parentheses.

RESULTS

The constants given by the present measurements have been used to calculate the wavenumbers of the entire 10⁰0–02⁰0 band, which are given in Table IV. The wavenumbers for the 02⁰0–00⁰0 band are also determined from this work and from earlier frequency measurements and are given in Table V. Finally, the wavenumbers for the 02⁰1–00⁰0 band are determinable now from frequency measurements and are given in Table VI. To calculate these latter transitions we have also taken into account the *l*-type resonance between the 02⁰1 and 02²1 states as indicated above and in Ref. (4).

The present results are in very good agreement with the FTS measurements reported by Toth (7) for the 02⁰0–00⁰0 transition. The band centers differ by only 2 MHz (0.00006 cm⁻¹), and the rotational constants are also in good agreement when allowance

TABLE V
Wavenumbers (in cm⁻¹) Calculated^a for the 02⁰0–00⁰0 Band of N₂O

P-BRANCH	J''	R-BRANCH	P-BRANCH	J''	R-BRANCH
1167.29433(5)	0	1168.97219(5)	1135.90939(5)	40	1203.80794(5)
1166.45813(5)	1	1169.81384(5)	1135.13073(5)	41	1204.69940(5)
1165.62376(5)	2	1170.65730(5)	1134.35286(5)	42	1205.59122(5)
1164.79121(5)	3	1171.50255(5)	1133.57574(5)	43	1206.48332(5)
1163.96048(5)	4	1172.34958(5)	1132.79933(5)	44	1207.37567(5)
1163.13158(5)	5	1173.19838(5)	1132.02358(5)	45	1208.26822(5)
1162.30449(5)	6	1174.04894(5)	1131.24845(5)	46	1209.16091(5)
1161.47922(5)	7	1174.90124(5)	1130.47390(5)	47	1210.05370(5)
1160.65574(5)	8	1175.75527(5)	1129.69989(5)	48	1210.94654(5)
1159.83407(5)	9	1176.61099(5)	1128.92638(5)	49	1211.83938(5)
1159.01417(5)	10	1177.46840(5)	1128.15332(5)	50	1212.73218(5)
1158.19605(5)	11	1178.32747(5)	1127.38069(5)	51	1213.62488(5)
1157.37968(5)	12	1179.18817(5)	1126.60844(5)	52	1214.51745(5)
1156.56506(5)	13	1180.05048(5)	1125.83654(5)	53	1215.40985(5)
1155.75215(5)	14	1180.91437(5)	1125.06495(5)	54	1216.30202(5)
1154.94095(5)	15	1181.77982(5)	1124.29363(5)	55	1217.19393(5)
1154.13144(5)	16	1182.64678(5)	1123.52256(5)	56	1218.08555(5)
1153.32358(5)	17	1183.51524(5)	1122.75169(5)	57	1218.97682(5)
1152.51737(5)	18	1184.38515(5)	1121.98100(5)	58	1219.86771(5)
1151.71276(5)	19	1185.25649(5)	1121.21045(5)	59	1220.75819(5)
1150.90974(5)	20	1186.12920(5)	1120.44003(5)	60	1221.64822(6)
1150.10828(5)	21	1187.00327(5)	1119.66969(5)	61	1222.53776(6)
1149.30835(5)	22	1187.87864(5)	1118.89941(5)	62	1223.42679(6)
1148.50991(5)	23	1188.75528(5)	1118.12917(5)	63	1224.31526(6)
1147.71294(5)	24	1189.63314(5)	1117.35895(6)	64	1225.20316(6)
1146.91741(5)	25	1190.51219(5)	1116.58871(6)	65	1226.09044(6)
1146.12327(5)	26	1191.39238(5)	1115.81843(6)	66	1226.97709(6)
1145.33050(5)	27	1192.27366(5)	1115.04810(6)	67	1227.86306(6)
1144.53905(5)	28	1193.15599(5)	1114.27769(6)	68	1228.74835(6)
1143.74889(5)	29	1194.03932(5)	1113.50719(6)	69	1229.63291(6)
1142.95998(5)	30	1194.92360(5)	1112.73658(6)	70	1230.51672(6)
1142.17228(5)	31	1195.80879(5)	1111.96583(6)	71	1231.39977(7)
1141.38576(5)	32	1196.69483(5)	1111.19493(6)	72	1232.28202(8)
1140.60036(5)	33	1197.58168(5)	1110.42387(7)	73	1233.16345(9)
1139.81605(5)	34	1198.46929(5)	1109.65262(7)	74	1234.04404(10)
1139.03279(5)	35	1199.35760(5)	1108.88119(8)	75	1234.92377(12)
1138.25054(5)	36	1200.24656(5)	1108.10954(10)	76	1235.80262(14)
1137.46925(5)	37	1201.13613(5)	1107.33767(11)	77	1236.68056(16)
1136.68888(5)	38	1202.02625(5)	1106.56557(13)	78	1237.55757(19)
	39	1202.91687(5)	1105.79321(16)	79	1238.43364(23)

a) The uncertainty in the last digits (one standard error) is given in parentheses.

TABLE VI
Wavenumbers (in cm^{-1}) Calculated^a for the 02⁰1-00⁰ Band of N₂O

P-BRANCH	J''	R-BRANCH	P-BRANCH	J''	R-BRANCH
	0	2462.83261(6)	2436.07496(7)	30	2487.03460(7)
2461.15830(7)	1	2463.66718(6)	2435.18026(7)	31	2487.80701(7)
2460.31856(6)	2	2464.50000(6)	2434.28328(7)	32	2488.57681(7)
2459.47709(6)	3	2465.33108(6)	2433.38398(7)	33	2489.34392(7)
2458.63391(6)	4	2466.16040(6)	2432.48233(7)	34	2490.10832(7)
2457.78901(6)	5	2466.98794(6)	2431.57830(7)	35	2490.86996(8)
2456.94240(6)	6	2467.81370(6)	2430.67183(7)	36	2491.62877(8)
2456.09405(6)	7	2468.63767(6)	2429.76290(8)	37	2492.38473(9)
2455.24398(6)	8	2469.45981(6)	2428.85146(8)	38	2493.13777(9)
2454.39217(6)	9	2470.28012(6)	2427.93748(9)	39	2493.88786(10)
2453.53861(6)	10	2471.09857(6)	2427.02091(9)	40	2494.63494(11)
2452.68330(6)	11	2471.91515(6)	2426.10172(10)	41	2495.37897(12)
2451.82622(6)	12	2472.72983(6)	2425.17987(11)	42	2496.11990(13)
2450.96736(6)	13	2473.54258(6)	2424.25531(12)	43	2496.85768(13)
2450.10671(6)	14	2474.35338(6)	2423.32801(13)	44	2497.59226(15)
2449.24425(6)	15	2475.16220(6)	2422.39793(14)	45	2498.32359(16)
2448.37996(6)	16	2475.96902(6)	2421.46503(15)	46	2499.05164(17)
2447.51382(6)	17	2476.77380(6)	2420.52927(16)	47	2499.77635(18)
2446.64582(6)	18	2477.57651(6)	2419.59061(17)	48	2500.49767(20)
2445.77593(6)	19	2478.37712(6)	2418.64902(18)	49	2501.21557(22)
2444.90412(6)	20	2479.17560(7)	2417.70446(20)	50	2501.93000(25)
2444.03038(6)	21	2479.97190(7)	2416.75688(22)	51	2502.64091(28)
2443.15468(7)	22	2480.76600(7)	2415.80627(25)	52	2503.34826(31)
2442.27698(7)	23	2481.55786(7)	2414.85257(28)	53	2504.05202(36)
2441.39728(7)	24	2482.34743(7)	2413.89576(31)	54	2504.75213(41)
2440.51552(7)	25	2483.13467(7)	2412.93580(36)	55	2505.44856(47)
2439.63169(7)	26	2483.91956(7)	2411.97266(41)	56	2506.14127(54)
2438.74575(7)	27	2484.70203(7)	2411.00631(47)	57	2506.83023(62)
2437.85767(7)	28	2485.48206(7)	2410.03672(54)	58	2507.51538(71)
2436.96742(7)	29	2486.25960(7)	2409.06386(62)	59	2508.19671(81)

a) The uncertainty in the last digits (one standard error) is given in parentheses.

is made for the difference in the equations used to fit the transitions. Our Table V agrees quite closely with the values in Table 9 of Ref. (7).

ACKNOWLEDGMENTS

This work was supported in part by the NASA Office of Upper Atmospheric Research. We are grateful to D. A. Jennings for his generosity in providing the computer programming for the frequency synthesis.

RECEIVED: October 30, 1986

REFERENCES

1. C. R. POLLOCK, F. R. PETERSEN, D. A. JENNINGS, J. S. WELLS, AND A. G. MAKI, *J. Mol. Spectrosc.* **107**, 62-71 (1984).
2. J. S. WELLS, D. A. JENNINGS, A. HINZ, J. S. MURRAY, AND A. G. MAKI, *J. Opt. Soc. Amer. B* **2**, 857-861 (1985).
3. J. S. WELLS, A. HINZ, AND A. G. MAKI, *J. Mol. Spectrosc.* **114**, 84-96 (1985).
4. A. HINZ, J. S. WELLS, AND A. G. MAKI, *Z. Phys. D*, in press.
5. R. S. MULLIKEN, *J. Chem. Phys.* **23**, 1997-2011 (1967).

6. G. GUELACHVILI, *Canad. J. Phys.* **60**, 1334–1347 (1982).
7. R. A. TOTH, *J. Opt. Soc. Amer.* **B3**, 1263–1281 (1986).
8. C. AMIOT AND G. GUELACHVILI, *J. Mol. Spectrosc.* **59**, 171–190 (1976).
9. B. G. WHITFORD, K. J. SIEMSEN, H. D. RICCIUS, AND G. R. HANES, *Opt. Commun.* **14**, 70–74 (1975).
10. J. S. WELLS, F. R. PETERSEN, AND A. G. MAKI, *Appl. Opt.* **18**, 3567–3573 (1979).
11. C. FREED AND A. JAVAN, *Appl. Phys. Lett.* **17**, 53–56 (1970).
12. M. BOGEY, *J. Phys. B* **8**, 1934–1938 (1975).
13. F. J. LOVAS AND E. TIEMANN, *J. Chem. Phys. Ref. Data* **3**, 609–770 (1974).
14. K. JOLMA, J. KAUPPINEN, AND V.-M. HORNEMAN, *J. Mol. Spectrosc.* **101**, 278–284 (1983).
15. J. KAUPPINEN, private communication (Kauppinen's affiliation: Department of Physics, University of Oulu, Oulu, Finland).
16. A. G. MAKI AND D. R. LIDE, *J. Chem. Phys.* **47**, 3206–3210 (1967).