Heterodyne Frequency Measurements on N$_2$O near 1060 cm$^{-1}$

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The results of heterodyne frequency measurements are given for the 10$^0$0-02$^0$0 band of N$_2$O centered at 1056 cm$^{-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 $b$-type resonance between the 02$^0$0 and 02$^0$1 states. Derived tables of wavenumbers are given for the 02$^0$0-00$^0$0 band near 1168 cm$^{-1}$ and the 02$^0$1-00$^0$0 band near 2460 cm$^{-1}$.

INTRODUCTION

In order to provide accurate frequency calibration data using infrared transitions of N$_2$O (nitrous oxide), we have been making heterodyne frequency measurements on N$_2$O (1-4). In this paper we report frequency measurements on the very weak N$_2$O band centered near 1060 cm$^{-1}$ and due to the 10$^0$0−02$^0$0 transition. This is the band that is in weak Fermi resonance with the 10$^0$0−00$^0$1 band at 940 cm$^{-1}$ on which the N$_2$O laser is based.

We are not aware of any modern measurements of this band, but there are several good measurements of the 10$^0$0−00$^0$0 and 02$^0$0−00$^0$0 bands from which one can calculate the 10$^0$0−02$^0$0 band. Guelachvili (6) has given very good Fourier transform spectroscopy (FTS) measurements of the 02$^0$0−00$^0$0 band at 1168 cm$^{-1}$, but more recently Toth (7) has given more complete and (we believe) more accurate FTS measurements on the same band system. Aside from earlier measurements, the best direct measurements of the 10$^0$0−00$^0$0 transition near 2223 cm$^{-1}$ were made by Amiot and Guelachvili (8). The most accurate measurements involving the 10$^0$0 state are the heterodyne frequency measurements of the 10$^0$0−00$^0$1 transitions, which are the laser transitions of N$_2$O, by Whitford et al. (9). When combined with the heterodyne measurements on the 00$^0$1−00$^0$0 transitions reported by Wells et al. (3), the 10$^0$0 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 $v_1$ and $v_3$. 

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We have been measuring systematically the frequencies of many transitions of N$_2$O in an effort to determine the energy separation of all the lower states of N$_2$O 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 02$^0$ state to any of the other states, with the exception of a single frequency measurement of the $P(75)$ 02$^0$-00$^0$ transition (2). While the 10$^0$-02$^0$ 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 02$^0$ state with respect to the other low-lying vibrational states of N$_2$O.

Since the 10$^0$-02$^0$ transitions nicely overlap the region of the CO$_2$ 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 10$^0$-02$^0$ transitions against a well-characterized CO$_2$ laser. This in turn has enabled us to calculate accurate wavenumbers of the much stronger and therefore more useful 02$^0$-00$^0$ transitions near 1168 cm$^{-1}$. Furthermore, these data, when combined with the heterodyne measurements on the hot band transitions 02$^1$-02$^0$ given by Hinz et al. (4), allow us to calculate the wavenumbers of the 02$^0$-00$^0$ 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 CO$_2$ laser at a time was required for a heterodyne measurement. Most measurements used the normal $^{12}$C$_{16}$O$_2$ laser; a few measurements required a $^{13}$C$_{16}$O$_2$ 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 CO$_2$ lasers is about $1 \times 10^{-9}$, which is negligible relative to the other measurement uncertainties.

The basic technique was to lock the tunable diode-laser (TDL) frequency to the N$_2$O feature of interest and then heterodyne the TDL output with a nearby CO$_2$ laser transition in a HgCdTe mixer. For practical purposes, the measurement uncertainty was taken as one-tenth of the TDL-CO$_2$-laser beat-note linewidth plus a contribution due to the TDL lock scheme. This contribution was the Doppler width of the N$_2$O 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 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 state), the present measurements were combined with other infrared and microwave measurements involving the same state. Because there is a rather strong I-type resonance between the 02°0 and 02°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°0-02°0 Band of Nitrous Oxide

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
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</thead>
<tbody>
<tr>
<td>12Cl16O2</td>
<td>RII(28)</td>
<td>648.2 MHz</td>
<td>P(20) 31 094 136.7(40)</td>
</tr>
<tr>
<td>12Cl16O2</td>
<td>RII(24)</td>
<td>-2554.4 MHz</td>
<td>P(14) 31 270 692.8(40)</td>
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<tr>
<td>12Cl16O2</td>
<td>RII(22)</td>
<td>-1440.4 MHz</td>
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<td>+403.1 MHz</td>
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<td>RII(14)</td>
<td>+512.8 MHz</td>
<td>P(4) 31 546 541.7(40)</td>
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<td>+2582.6 MHz</td>
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<td>12Cl16O2</td>
<td>RII(30)</td>
<td>-1317.7 MHz</td>
<td>R(44) 32 515 225.9(70)</td>
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</tbody>
</table>

*a* The uncertainty in the last digits is given in parentheses.
TABLE II

$\text{N}_2\text{O} \text{ Rotational Constants}^a \left(\text{in cm}^{-1}\right) \text{ Used in the Present Analysis}$

<table>
<thead>
<tr>
<th>Vib. State</th>
<th>$B$</th>
<th>$D_{10}^7$</th>
<th>$E_{10}^{13}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>00$^2\Pi_0$</td>
<td>0.419010993(16)</td>
<td>1.760727(170)</td>
<td>-0.1787(324)</td>
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<tr>
<td>10$^2\Pi_0$</td>
<td>0.415559533(19)</td>
<td>1.754940(275)</td>
<td>-0.084(73)</td>
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<tr>
<td>02$^2\Pi_0$</td>
<td>0.419519782(24)</td>
<td>1.869710(406)</td>
<td>-1.547(189)</td>
</tr>
<tr>
<td>02$^2\Pi_1$</td>
<td>0.420124835(19)</td>
<td>1.817151(364)</td>
<td>1.016(253)</td>
</tr>
<tr>
<td>02$^2\Pi_1$</td>
<td>0.41814807(51)</td>
<td>1.89653(242)</td>
<td></td>
</tr>
<tr>
<td>02$^2\Pi_2$</td>
<td>0.418529986(80)</td>
<td>1.754197(793)</td>
<td>2.292(217)</td>
</tr>
<tr>
<td>02$^2\Pi_3$</td>
<td>0.41815607(51)</td>
<td>1.89653(242)</td>
<td></td>
</tr>
<tr>
<td>02$^2\Pi_4$</td>
<td>0.41784807(51)</td>
<td>1.89653(242)</td>
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</tr>
<tr>
<td>02$^2\Pi_5$</td>
<td>0.417529986(80)</td>
<td>1.754197(793)</td>
<td>2.292(217)</td>
</tr>
</tbody>
</table>

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

For the 02$^2\Pi_0$ state $\Delta\lambda=-1.99(40) \times 10^{-1} \text{ cm}^{-1}$ was also determined.

$c)$ The value of $G_z - G_x = 12.80221 \text{ cm}^{-1}$ was used for the calculation of the $l$-type resonance between the 02$^2\Pi_1$ and 02$^2\Pi_1$ levels.

The $10^0\Pi_0$ energy levels, which are the upper state of the present measurements, were calculated in the least-squares fit by the equation

$$E = G_x + B_x(J + 1) - D_x[J(J + 1) - l^2] + H_x[J(J + 1) - l^2],$$

(1)

In the analysis of the lower state (02$^0\Pi_0$) we used the following equations (16) in order to include the effect of the $l$-type resonance with the 02$^0\Pi_0$ state,

$$E(02^0\Pi_0) = E_0^0 + \frac{1}{2} \delta + \frac{1}{4} \{ \delta^2 + 4q^2[J^2(J + 1)^2 - 2J(J + 1)] \}^{1/2},$$

(2)

$$E(02^2\Pi_0) = E_0^0,$$

(3)

and

$$E(02^2\Pi_0) = E_0^0 - \frac{1}{2} \delta + \frac{1}{4} \{ \delta^2 + 4q^2[J^2(J + 1)^2 - 2J(J + 1)] \}^{1/2},$$

(4)

TABLE III

$\text{Band Centers Determined for N}_2\text{O}$

<table>
<thead>
<tr>
<th>Vib. Transitions</th>
<th>$\nu_0(\text{cm}^{-1})$</th>
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<tr>
<td>10$^0\Pi_0$-02$^0\Pi_0$</td>
<td>1053.624325(68)</td>
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<td>02$^0\Pi_0$-00$^0\Pi_0$</td>
<td>1168.132354(96)</td>
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<tr>
<td>02$^1\Pi_0$-02$^0\Pi_0$</td>
<td>1293.863964(88)</td>
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<td>02$^1\Pi_1$-00$^0\Pi_0$</td>
<td>2461.996319(130)</td>
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<tr>
<td>10$^0\Pi_0$-00$^0\Pi_0$</td>
<td>2223.756680(74)</td>
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</tbody>
</table>

$a)$ The uncertainty in the last digits (twice the standard error) is given in parentheses.
where $E_0$ and $E_0'$ are given by Eq. (1), with $l = 0$ and 2, respectively,

$$\delta = E_0 - E_0', \quad (5)$$

and

$$q = q_0 - q_{zz} R(J + 1) + q_{uJ} F(J + 1)^2. \quad (6)$$

The observed transitions are then given by

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

and

$$\nu_0 = G' - G''. \quad (8)$$

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**

<table>
<thead>
<tr>
<th>P-BRANCH</th>
<th>J''</th>
<th>R-BRANCH</th>
<th>P-BRANCH</th>
<th>J''</th>
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<td>1054.78469(3)</td>
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<td>1073.53950(19)</td>
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</table>

a) The uncertainty in the last digits (one standard error) is given in parentheses.
The constants given by the present measurements have been used to calculate the wavenumbers of the entire $^{10}$$^{0}$$^{0}$$^{0}$--$^{0}$$^{2}$$^{0}$$^{0}$ band, which are given in Table IV. The wavenumbers for the $^{0}$$^{2}$$^{0}$$^{0}$--$^{0}$$^{0}$$^{0}$ band are also determined from this work and from earlier frequency measurements and are given in Table V. Finally, the wavenumbers for the $^{0}$$^{2}$$^{1}$--$^{0}$$^{0}$$^{0}$ band are determinable now from frequency measurements and are given in Table VI. To calculate these latter transitions we have also taken into account the $i$-type resonance between the $^{0}$$^{2}$$^{1}$ and $^{0}$$^{2}$$^{1}$ 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 $^{0}$$^{2}$$^{0}$$^{0}$--$^{0}$$^{0}$$^{0}$ 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 $^{0}$$^{2}$$^{0}$--$^{0}$$^{0}$$^{0}$ Band of N$_{2}$O |
|---|---|---|---|---|
| P-BRANCH | J$^a$ | R-BRANCH | P-BRANCH | J$^a$ |
| 1167.29433(5) | 1 | 1169.81384(5) | 1135.90093(5) | 40 |
| 1166.45813(5) | 2 | 1170.65730(5) | 1134.35286(5) | 42 |
| 1165.62736(5) | 3 | 1171.50235(5) | 1133.57574(5) | 43 |
| 1164.79121(5) | 4 | 1172.34958(5) | 1132.79933(5) | 44 |
| 1163.96048(5) | 5 | 1173.19838(5) | 1132.02538(5) | 45 |
| 1163.13158(5) | 6 | 1174.04894(5) | 1131.24845(5) | 46 |
| 1162.30449(5) | 7 | 1174.90124(5) | 1130.47390(5) | 47 |
| 1161.47922(5) | 8 | 1175.35527(5) | 1129.69589(5) | 48 |
| 1160.65574(5) | 9 | 1176.10999(5) | 1128.92638(5) | 49 |
| 1159.83407(5) | 10 | 1177.46840(5) | 1128.15332(5) | 50 |
| 1159.01417(5) | 11 | 1178.32747(5) | 1127.38069(5) | 51 |
| 1158.19065(5) | 12 | 1179.18171(5) | 1126.60844(5) | 52 |
| 1157.37968(5) | 13 | 1180.05048(5) | 1125.83654(5) | 53 |
| 1156.56506(5) | 14 | 1180.91437(5) | 1125.06495(5) | 54 |
| 1155.75213(5) | 15 | 1181.77982(5) | 1124.29363(5) | 55 |
| 1154.94095(5) | 16 | 1182.64678(5) | 1123.52256(5) | 56 |
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| 1145.33000(5) | 28 | 1193.15599(5) | 1114.27769(5) | 68 |
| 1144.53905(5) | 29 | 1194.03932(5) | 1113.50719(5) | 69 |
| 1143.74899(5) | 30 | 1194.92360(5) | 1112.73658(5) | 70 |
| 1142.95998(5) | 31 | 1195.80879(5) | 1111.96583(5) | 71 |
| 1142.17223(5) | 32 | 1196.64543(5) | 1111.19493(5) | 72 |
| 1141.38576(5) | 33 | 1197.51218(5) | 1110.42337(5) | 73 |
| 1140.60036(5) | 34 | 1198.46929(5) | 1109.65262(5) | 74 |
| 1139.81605(5) | 35 | 1199.35760(5) | 1108.88119(5) | 75 |
| 1139.03279(5) | 36 | 1200.24656(5) | 1108.10954(5) | 76 |
| 1138.25054(5) | 37 | 1201.13613(5) | 1107.33767(11) | 77 |
| 1137.46925(5) | 38 | 1202.02625(5) | 1106.56557(13) | 78 |
| 1136.68888(5) | 39 | 1202.91687(5) | 1105.79321(16) | 79 |
| | | | | |

$^a$ The uncertainty in the last digits (one standard error) is given in parentheses.
### Table VI

Wavenumbers (in cm⁻¹) Calculated for the 021°1-0000 Band of N₂

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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.

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REFERENCES

15. J. Kauppinen, private communication (Kauppinen’s affiliation: Department of Physics, University of Oulu, Oulu, Finland).