HETERODYNE DETECTION OF THE 752.033-GHz H$_2$O ROTATIONAL ABSORPTION LINE

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SUMMARY

A tunable high-resolution two-stage heterodyne radiometer has been developed for the purpose of investigating the intensity and lineshape of the 752.033-GHz (2$_{11}$ + 2$_{02}$) rotational transition of water vapor. Single-sideband system noise temperatures of approximately 45,000 K have been obtained using a sensitive GaAs Schottky diode as the first-stage mixer. First local-oscillator power was supplied by a CO$_2$-laser pumped formic acid laser (761.61 GHz), generating an X-band IF signal with theoretical line center at 9.5744 GHz. Second local-oscillator power was provided by a tunable C-band source. A resolution capability of 1 MHz was achieved by means of a 3-GHz waveguide cavity filter with only 9-dB insertion loss.

In absorption measurements of the H$_2$O taken from a laboratory simulation of a high-altitude rocket plume, the center frequency of the 752-GHz line was determined to within 1 MHz of the reported value. A rotational temperature ~75 K, a linewidth ~5 MHz and a Doppler shift ~3 MHz (from a 45-degree rotation of the flow direction) were measured with the line-of-sight intersecting the simulated-plume axis at a distance downstream of 30 nozzle diameters. These absorption data were obtained against continuum background radiation sources at temperatures of 1175 and 300 K.

INTRODUCTION

Recent advances in mixer technology at submillimeter-wavelengths have made possible the development of heterodyne receivers with single-sideband system temperatures below 20,000 K (ref. 1). As part of a Lincoln Laboratory program to investigate the submillimeter-wave radiative properties of H$_2$O molecules in high-altitude rocket plumes (refs. 1,2), a heterodyne receiver similar to that reported earlier (ref. 1) has been adapted to measure the rotational temperature of the 752.033 GHz (ref. 3) transition (2$_{11}$ + 2$_{02}$) of H$_2$O (a prominent plume constituent) under simulated conditions of high altitude (~150 km). At altitudes where collisions with ambient molecules are sufficiently infrequent, adiabatic cooling through rapid volume expansion of H$_2$O vapor emitted into the

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vacuum from a nozzle can produce rotational temperatures of selected transitions well below 100 K (refs. 2,4). In work reported recently, Bulatov et al. (ref. 5) performed active measurements with a spectrometer on four submillimeter-wave H_2O lines (including the 2_{11} \rightarrow 2_{02} transition) in a planar stream under conditions of relatively high ambient pressure through a point almost at the exit plane. This paper describes the experimental details and some initial results of passive measurements of H_2O emitted from a simulated rocket into a high vacuum at a point farther downstream (ref. 6).

THEORETICAL CONSIDERATIONS

Based on the results of a theoretical analysis (refs. 2,4), it has been concluded that the combination of high optical depth and low rotational temperature in a water vapor vacuum expansion would most likely occur for the low-lying rotational transitions, particularly the 1_{10} \rightarrow 1_{01} at \approx 557 GHz and the 2_{11} \rightarrow 2_{02} at \approx 752 GHz (fig. 1). At these spectral-line frequencies, temperatures far below room temperature (\approx 50 K) are expected to be reached within a short distance from the nozzle through adiabatic cooling by volume expansion.

With the conditions of low pressure and temperature anticipated in these experiments, pressure broadening or self-broadening of the spectral line is not possible in the downstream regime of the plume. Thermal Doppler effects from random molecular motion would normally be the dominant broadening mechanism, and would be determined by

$$\Delta v_{\text{thermal}} = 2\left(\frac{2kT \ln 2}{mc^2}\right)^{1/2} v,$$

where m is the mass of the H_2O molecule, k is Boltzmann's constant, and c is the velocity of light. Since \Delta v_{\text{thermal}} = 2.2 MHz at v = 752 GHz and T = 300 K, however, it can be argued that the principal source of line broadening in the expanding plume is more likely to be the molecular flow velocities.

Because the molecular flow velocity v_{flow} of the plume can reach several km/sec, its Doppler effects are expected not only to cause line-center frequency shifts \Delta v_{\text{shift}} which depend on the direction of the flow relative to the observer, but also to control the linewidth \Delta v_{\text{width}}, which is a function of v_{flow} and the plume divergence angle. In figure 2, an elementary model depicting these Doppler effects is sketched. With the flow directed at an angle \Theta to the observer, the frequency shift may be estimated by the relation

$$\Delta v_{\text{shift}} \approx \frac{v_{\text{flow}}}{c} v \sin \Theta.$$

For a plume divergence angle \alpha (defining the optically thick part of the plume), the Doppler linewidth may be estimated by
\[ \Delta \nu_{\text{width}} \sim \frac{2v_{\text{flow}}}{c} \nu \sin(\alpha/2). \]  

(3)

With \( \alpha = \pi/2 \), and \( v_{\text{flow}} = 2 \text{ km/sec} \), for example, \( \Delta \nu_{\text{width}} \sim 7 \text{ MHz} \) at \( \nu = 752 \text{ GHz} \), a linewidth significantly greater than even the room temperature value of the thermal Doppler width estimated above. For a cold plume temperature (\( \approx 75 \text{ K} \)), the \( \Delta \nu_{\text{thermal}} \) is reduced to 1.1 MHz.

EXPERIMENTAL TECHNIQUES

To measure the rotational temperature of the 752-GHz line, a high-resolution heterodyne radiometer was employed. As shown in figure 3, a formic acid (HCOOH) FIR laser, pumped by a 40-W CO\(_2\) laser at 10.6 \( \mu \text{m} \) was used to provide local oscillator power for the first-heterodyne stage of the radiometer. The strong line at 761.6 GHz is capable of delivering 30 mW and is within 10 GHz of the H\(_2\)O rotational line of interest. First-stage mixing was accomplished by means of a GaAs Schottky diode (ref. 1). After intermediate frequency (IF) amplification with a low-noise X-band FET amplifier, the signal was mixed down to 8-band by means of a C-band sweep generator to provide an IF for which a 0.9-MHz bandwidth, 3-GHz cavity waveguide filter with only 9-dB insertion loss was utilized (fig. 4). Because the linewidths of thermally broadened rotational lines were on the order of only a few MHz, the importance of a filter capable of providing resolution less than 1 MHz is crucial in any application where molecular densities and optical depths are low enough to produce linewidths that approach the thermal Doppler limit. Another feature of this two-stage receiver was the continuously fine tunability afforded by the second local oscillator.

To make passive measurements of narrow-line spectra with a heterodyne receiver, it is important to remember that, while two sidebands are generated by the mixing process, only one of them will contain the desired information. In figure 5, it is explained how the two sidebands combine in the first IF stage. In the case depicted, a room temperature absorption line in the lower sideband is observed against a hot blackbody continuum, with the baseline for each sideband supplied by the room temperature chopper blade reference. Since the upper sideband is not filtered out, the baseline of the lower sideband must be identified as the top of the upper sideband contribution when the two sidebands are combined.

A convenient feature of this particular experiment lies in the fact that collision broadening of the atmospheric H\(_2\)O line is large enough to filter out the entire lower sideband when steps to remove this moisture from the optical path are not taken. As a result, the upper sideband signal contribution can be isolated and recorded initially, and then the volume surrounding optical path of the signal purged in a dry N\(_2\) enclosure (fig. 4) to permit the passage of the lower sideband containing the absorption line. Once this is accomplished,
the resultant signal is doubled (except over the narrow band of the H₂O absorption line) and the measurements can proceed, using the top of the upper sideband as the 300-K reference. By measuring the signal from a liquid-nitrogen-cooled (77 K) blackbody source, the output voltage from the phase-sensitive detector (PSD) can be calibrated directly in terms of temperature. With this calibration source, a single-sideband system noise temperature of 45,000 K was determined.

For the radiometric observations of simulated rocket plumes, a large high-vacuum chamber (≈ 10 m long by 3 m in diameter) with a liquid-nitrogen cryopanned lining was used to create the conditions of high altitude. Along the axis of the chamber, a laboratory-scale jet with a sonic nozzle was used to generate a controlled flow of adiabatically expanding H₂O molecules, as shown in figure 6. To provide a background of temperature greater than that of the reference, in this case a 300-K chopper blade, a mercury-arc lamp was installed at a port opposite to the Z-cut quartz exit window. By defocussing the collimating optics (fig. 7), a real image of the source may be established at the first-stage mixer, with the ray paths parallel to the beam from the FIR local oscillator. With this background, a signal-to-rms noise of ≈ 20 was attainable for an integration time of 4 seconds.

EXPERIMENTAL RESULTS

With the chamber evacuated to a roughing pressure of 10⁻² Torr and the arc lamp turned on, an optically thick absorption of the 2₁₁⁻→₂₀₂ transition was measured with center at 9.5737 ± 0.0010 GHz in the first IF band (table I). In figure 8, the trace of the Lorentzian-shaped line is presented, with a scale determined by calibrating the radiometer by means of a 77 K blackbody (a sheet of absorbing material cooled by liquid nitrogen). The mercury-arc lamp temperature was measured at 1000 K, and raised to 1175 K after correcting for the effect of the 300-K quartz window with emissivity ≈ 0.20. The postdetection integration time for these measurements was 4 seconds. If the frequency of the formic acid LO line is taken as 761.6076 ± 0.0005 GHz (ref. 7) and the H₂O line frequency as 752.0332 ± 0.0005 GHz (ref. 3), the expected first IF is 9.5744 ± 0.0010 GHz, less than 1 MHz from the measured value. The discrepancy between these two values can be accounted for by the accumulated measurement errors involved in the present experiment and the referenced work. The minimum linewidth observed was approximately 2.5 MHz, in good agreement with the calculated thermal (Doppler) width of 2.2 MHz for H₂O at 300 K.

With the chamber walls cooled to 77 K and the pressure reduced to ≈ 10⁻⁶ Torr, the rotational temperature of the H₂O line in the plume of the steam jet was investigated. For a water flow rate of 1 g/sec and a stagnation temperature of 900 K, the theoretical prediction (ref. 4) of the signal brightness or antenna temperature Tₐ as a function of distance downstream (in terms of nozzle diameters R) along the plume axis is given by the curve in figure 9. In figure 10, the absorption line trace for R = 30 is presented. The temperature at the line center was 75 K (after applying the quartz window emissivity correction to the measured value of 120 K), and the linewidth was 5.5 MHz.
In figure 11, the results of a second experiment are presented. To test for the occurrence of a Doppler shift, the steam jet was rotated away from the radiometer through an angle $\theta = \pi/4$ and the absorption measurement repeated. In this case, the line center frequency was red-shifted by 3 MHz, although its width decreased to 4.2 MHz because of the likelihood that a portion of the plume was striking the cold wall before passing through the radiometer beam. Finally, the absorption was observed with a 300-K background (i.e., with the lamp turned off) and the temperature and linewidth were in agreement with the data taken for the hot background.

The results of frequency, linewidth and Doppler shift measurements are listed in table I. From equation (3), an effective angle $\alpha = 80^\circ$ and a flow velocity $v_{\text{flow}} = 1.7 \text{ km/sec}$ (ref. 8) would provide a linewidth of 5.5 MHz. With the plume directed away from the radiometer ($\theta = \pi/4$), and with wall effects taken into account, the calculated value of $\Delta \nu_{\text{width}}$ for the same plume conditions was 3.8 MHz, in reasonable agreement with the measured value of 4.2 MHz mentioned above. The calculated estimate of $\Delta \nu_{\text{shift}}$ from equation (2) for these plume conditions was 3.0 MHz, in agreement with the measured value.

CONCLUSIONS

With the aid of a high-resolution two-stage heterodyne radiometer, spectral absorption measurements of the 752.033 GHz line of H$_2$O vapor were carried out using a blackbody continuum as a background radiation source for investigating the absorptive properties of the H$_2$O content of simulated high altitude rocket plumes. The receiver had a resolution capability of 1 MHz and was tunable through the use of a C-band microwave sweep generator as a second-stage local-oscillator source. With thermally Doppler broadened H$_2$O at room temperature, the line center frequency was determined within 1 MHz of the reported value, and the linewidth was in close agreement with the calculated value.

To simulate the physical situation of a high altitude rocket in a laboratory environment, a small steam jet was operated inside a large high vacuum chamber, with the H$_2$O jet plume traversing the radiometer line-of-sight. These experiments verified that this rotational line is optically thick, with excitation temperatures below 100 K, in the downstream part of the plume, as predicted by theoretical modeling. From the measured traces of the absorption line profile, it was concluded that Doppler effects from plume molecular flow velocity components along the radiometer line-of-sight were the principal sources of the measured linewidth.

A Doppler shift in the line center frequency which occurred when the plume direction was rotated relative to the line-of-sight was also judged to be caused by the change in direction of the molecular flow velocity component along the plume axis, as verified by the agreement between the experimental result and theoretical prediction.

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technology required to carry out these measurements, to C. D. Parker for assistance in the operation of the heterodyne receiver, and to W. E. Courtney for providing the narrow-band filter. This work was sponsored by the Department of the Army.
REFERENCES


TABLE I.-SUMMARY OF EXPERIMENTAL RESULTS FOR $R = 30$, $v_{flow} = 1.7$ km/sec, and $\alpha = 80^\circ$

<table>
<thead>
<tr>
<th>First IF (GHz)</th>
<th>Measured</th>
<th>Calculated</th>
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<tbody>
<tr>
<td></td>
<td>$9.5737 \pm 0.0010$</td>
<td>$9.5744 \pm 0.0010$</td>
</tr>
<tr>
<td>Plume Temperature (K)</td>
<td>75</td>
<td>80</td>
</tr>
<tr>
<td>$\Delta\nu_{width}$ with $\theta = 0$ (MHz)</td>
<td>5.5</td>
<td>5.5</td>
</tr>
<tr>
<td>$\Delta\nu_{width}$ with $\theta = \pi/4$ (MHz)</td>
<td>4.0</td>
<td>3.8</td>
</tr>
<tr>
<td>$\Delta\nu_{width}$ for $\theta = \pi/4$ (MHz)</td>
<td>3.0</td>
<td>3.0</td>
</tr>
</tbody>
</table>
Figure 1. Water molecule rotational energy levels.
\[ \Delta \nu_{\text{shift}} \approx \frac{v_{\text{flow}}}{c} \nu \sin \theta \]

\[ \Delta \nu_{\text{width}} \approx \frac{2v_{\text{flow}}}{c} \nu \sin (\alpha/2) \]

Figure 2.- Elementary model section of plume expansion into a vacuum, defining the plume angle \( \alpha \) and the plume direction angle \( \theta \) relative to the radiometer line-of-sight.

Figure 3.- Optical arrangement for submillimeter-wave (FIR) laser with CO\(_2\) laser pump source.
Figure 4.- Block diagram of two-stage heterodyne radiometer for operation at 752 GHz, with high-vacuum space chamber and water vapor jet.

Figure 5.- Single-sideband detection of a narrow absorption line in the presence of two sidebands, using a hot continuum background source.
Figure 6. - Laboratory-scale water vapor jet with sonic nozzle.

Figure 7. - Optical arrangement for the mercury-arc lamp, paraboloidal mirror and Schottky-barrier diode mixer.
Figure 8. - Experimental trace of thermally broadened H₂O line at 752.033 GHz with vacuum chamber walls at 300 K.

Figure 9. - Theoretical prediction of jet plume brightness temperature in laboratory rocket simulation experiment as a function of R (nozzle diameters downstream).
Figure 10.- Experimental trace of plume H$_2$O line at 752.033 GHz with vacuum chamber walls at 77 K and for $R = 30$ and $\theta = 0$.

Figure 11.- Experimental trace of H$_2$O line at 752.033 GHz with vacuum chamber walls at 77 K for the case of $R = 30$ and $\theta = \pi/4$, showing the Doppler shift. Data are presented for both 1175 K and 300 K continuum background cases.