LABORATORY MEASUREMENTS OF THE PURE ROTATION S(2) AND S(3) TRANSITIONS IN H$_2$

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ABSTRACT

Frequencies and transition rates have been measured for S(2) and S(3) in the pure rotation ground-state spectrum of molecular hydrogen. For S(2) the determined frequency, 814.4250 ± 0.0005 cm$^{-1}$, is 0.027 cm$^{-1}$ lower than the value used by Beck, Lacy, and Geballe in their detection of this line in the Orion molecular cloud. The S(3) frequency is 1034.67035 ± 0.00010 cm$^{-1}$. Transition probabilities measured for *S'(2) and *S(3) are $3.1 \pm 0.4 \times 10^{-9}$ s$^{-1}$ and $9.9 \pm 0.5 \times 10^{-9}$ s$^{-1}$, respectively.

Subject headings: interstellar: molecules — laboratory spectra — transition probabilities

I. INTRODUCTION

The infrared quadrupole spectrum of molecular hydrogen has gained considerable astrophysical importance since the detection (Gautier et al. 1976) in the Orion molecular cloud of seven vibration-rotation 1–0 lines near 2 μm. Pure rotational 0–0 quadrupole spectra of H$_2$ were first successfully observed with the detection of emission from the 12 μm S(2) transition ($J = 4 \rightarrow 2$) in the Orion Nebula (Beck, Lacy, and Geballe 1979, hereafter BLG). More recently, Knacke and Young (1980, 1981) reported the detection in Orion of the high-$J$ transitions S(8), S(9), S(12), S(13), S(14), and S(15).

The velocities and source dynamics deduced from the Orion S(2) observations depend critically upon a knowledge of the rest frequency of that transition. An early determination of the frequency of the S(2) transition by Stoicheff (1957) from Raman spectroscopy had given 814.406 ± 0.020 cm$^{-1}$. When BLG used this value they found that it led to a 15 km s$^{-1}$ discrepancy between their 0–0 S(2) observations and the 1–0 H$_2$ line observations in Orion by Nadeau and Geballe (1979). To obtain a more accurate frequency for the S(2) transition, BLG instead combined the more recent Raman results of Cooper, May, and Gupta (1970) for S(0) and S(1) with the diode laser measurement for S(3) by Reid and McKellar (1978) and calculated 814.452 ± 0.005 cm$^{-1}$ for the S(2) frequency. This removed the apparent difference between the 0–0 and 1–0 emission velocities in Orion.

The large deviation of the Stoicheff measurement of S(2) from the derivation of BLG, as well as the importance in the Orion observations of knowing the correct frequency for this transition, have prompted a new laboratory investigation of the pure rotation spectrum of molecular hydrogen.

II. EXPERIMENT

Laboratory spectra of the S(2) and S(3) transitions were taken using the Fourier transform spectrometer (FTS) of the McMath telescope at Kitt Peak. The unapodized spectral resolution was 0.005 cm$^{-1}$. The detector was liquid helium cooled, arsenic-doped silicon, and the beamsplitters were KBr.

The sample was contained in a White multiple-traversal cell set for 433.96 m total path length. Sample pressure was measured with a Bourdon-type Heise gauge and corrected for ambient atmospheric pressure. The sample temperature was 296 K. The spectral frequency calibration was checked by recording NH$_3$ and H$_2$O spectra simultaneously with the H$_2$ spectra; these were subsequently recorded again with CO$_2$ in the White cell to provide standard frequencies (CO$_2$ frequencies were taken from Petersen et al. 1974).

Both a 450 watt glower continuum source and a tunable diode laser were used as infrared sources during the experiment. The continuum source yielded broadband spectra at 2.61 atm pressure which allowed us to record the S(2) and S(3) lines simultaneously. The diode laser was used for measurements of S(2) in the range 0.3–2.7 atm and was modulated (at 3–5 kHz, a rate faster than the FTS sampling rate) in an ~ 0.1 cm$^{-1}$ interval containing the hydrogen transition. The glower
and laser spectra gave consistent results, and no measurable pressure-shift was found. Conventional diode laser spectra were also recorded to fully resolve the line shape for use in measuring line intensities.

The $aP(6,3)$ transition in $v_2$ of NH$_3$ was used by BLG to calibrate their Orion spectra. To eliminate this possible source of error in the H$_2$ observations, we also measured the frequency of this transition: $814.2416 \pm 0.0005$ cm$^{-1}$.

III. RESULTS AND DISCUSSION

Figure 1 shows the laboratory spectra of the $S(2)$ transition, from which the transition frequency was found to be $814.4250 \pm 0.0005$ cm$^{-1}$. The $S(3)$ transition frequency was $1034.67035 \pm 0.00010$ cm$^{-1}$. Measured frequencies and transition probabilities are compared with previous determinations in Table 1. Although of higher accuracy, the present result for $S(3)$ agrees very well with the diode laser measurement of Reid and McKellar (1978). The earlier Raman measurements by Stoicheff (1957) of $S(2)$ and $S(3)$ are both different from ours by $-0.019$ cm$^{-1}$, which is close to the error limit quoted in that work and may indicate a systematic experimental shift. In view of the present results, a redetermination of the $S(0)$ and $S(1)$ frequencies (measured previously by Stoicheff and more recently by Cooper, May, and Gupta 1970) is probably also warranted.

Our measured $S(2)$ frequency is lower by $0.027$ cm$^{-1}$ than that used by BLG in their observations of the Orion molecular cloud. This difference is offset somewhat by our corrected frequency for the $aP(6,3)$ transition in $v_2$ of NH$_3$, which was used by BLG to calibrate their spectrometer. Our measured value for this transition frequency, $814.2416 \pm 0.0005$ cm$^{-1}$, is lower by $0.008$ cm$^{-1}$ than the value used by BLG (taken from Garing, Nielsen, and Rao 1959). This calibration correction in turn reduces the frequency of the observed $S(2)$ emission in Orion by $0.008$ cm$^{-1}$. Combining this with our new frequency for $S(2)$ places the $S(2)$ peak emission in Orion near $0$ km s$^{-1}$ LSR velocity instead of near $+7$ km s$^{-1}$ as reported by BLG. (Note that a recent measurement of the NH$_3$ line frequency, reported by Urban et al. 1980, gave $814.239 \pm 0.005$ cm$^{-1}$.)

The 2 $\mu$m molecular hydrogen lines peak near the 9 km s$^{-1}$ LSR velocity of the extended molecular cloud in Orion (Nadeau and Geballe 1979; BLG). The 12 $\mu$m, H$_2$ emission peak velocity may also be consistent with 9 km s$^{-1}$ to within the limitations of resolution.

<table>
<thead>
<tr>
<th>Assignment</th>
<th>Frequency (cm$^{-1}$)</th>
<th>Transition Probability (s$^{-1}$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>$S(2)$</td>
<td>814.4250(5)$^a$</td>
<td>$3.1(4) \times 10^{-9}$</td>
<td>This work</td>
</tr>
<tr>
<td>$S(2)$</td>
<td>814.452(5)</td>
<td>...</td>
<td>BLG</td>
</tr>
<tr>
<td>$S(2)$</td>
<td>814.406(20)</td>
<td>...</td>
<td>Stoicheff 1957</td>
</tr>
<tr>
<td>$S(3)$</td>
<td>1034.67035(10)</td>
<td>$9.9(5) \times 10^{-9}$</td>
<td>This work</td>
</tr>
<tr>
<td>$S(3)$</td>
<td>1034.6702(7)</td>
<td>$9.7(4) \times 10^{-9}$</td>
<td>Reid and McKellar 1978$^b$</td>
</tr>
<tr>
<td>$S(3)$</td>
<td>1034.651(20)</td>
<td>...</td>
<td>Stoicheff 1957</td>
</tr>
</tbody>
</table>

$^a$Errors in parentheses are in units of the least significant digit.

$^b$Transition rate calculated from reported line strength.
(44 km s\(^{-1}\)) and signal to noise in the data reported by BLG. If the shift to 0 km s\(^{-1}\) is real, however, it would imply that the 0–0 and 1–0 emissions are probing different regions of the source. If the excitation is due to an expanding shell centered near the Becklin-Neugebauer object (Nadeau and Geballe 1979), the 12 \(\mu\)m peak emission region may be located in the blueshifted, near side of the shell. Alternate source models will yield different predictions, and the velocity centroid and shape of the emission may depend on extinction within the source. Definite conclusions will have to await higher signal-to-noise ratio spectra of Orion at 12 \(\mu\)m.

The strength of the \(S(2)\) line at 296 K was measured to be \(3.2 \pm 0.4 \times 10^{-3}\) cm\(^{-1}\) km\(^{-1}\) atm\(^{-1}\). This corresponds to a transition probability \(A = 3.1 \pm 0.4 \times 10^{-9}\) s\(^{-1}\), which is close to \(2.758 \times 10^{-9}\) s\(^{-1}\), the value calculated by Dalgarno and Wright (1972). The \(S(3)\) line strength was measured to be \(4.2 \pm 0.2 \times 10^{-3}\) cm\(^{-1}\) km\(^{-1}\) atm\(^{-1}\), yielding a transition probability \(A = 9.9 \pm 0.5 \times 10^{-9}\) s\(^{-1}\). This line strength agrees with that measured for \(S(3)\) by Reid and McKellar (1978), and the transition rate coincides with the calculation of Dalgarno and Wright (1972): \(9.850 \times 10^{-9}\) s\(^{-1}\). The transition rates for \(S(2)\) and \(S(3)\) also agree with the calculations of Turner, Kirby-Docken, and Dalgarno (1977), which are essentially identical to the Dalgarno and Wright (1972) results for these two transitions. The quadrupole matrix elements derived from these measurements for \(S(2)\) and \(S(3)\) are \(0.51 \pm 0.03\) \(ea^2\) and \(0.495 \pm 0.010\) \(ea^2\) (where \(e\) is the electron charge and \(a_0\) is the Bohr radius), again agreeing well with theoretical determinations (Birnbaum and Poll 1969; Dalgarno, Allison, and Browne 1969; Karl and Poll 1967).

The \(S(2)\) transition is the only low-\(J\) pure rotation line in molecular hydrogen which can be observed from the ground without severe masking by terrestrial absorption. It is our hope that these improved measurements will encourage additional observations of this transition in Orion and other sources.

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REFERENCES


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