ASSIGNMENT OF THE 1369.13 DIFFUSE INTERSTELLAR ABSORPTION BAND AND THREE OTHER FAR-ULTRAVIOLET INTERSTELLAR ABSORPTION LINES TO THE CH MOLECULE

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ABSTRACT

The three unidentified interstellar lines λ1368.74, 1369.13, and 1370.87 are here assigned to the CH molecule, including the diffuse interstellar band (DIB) at 1369.13 Å. A consistent rotational assignment of the interstellar lines and the 3d-X 2Π transition in laboratory spectrum is presented. The predominant broadening mechanism of the lines appears to be predissociation, which is found to be greater for lines with upper levels of d parity, including the DIB, than for lines with upper levels of c parity. The oscillator strength of the 3d-X 2Π transition is estimated to be f ≈ 0.033. Using the theoretical n* dependence of the parameters together with the wavenumber of the observed laboratory Q-form branch, an unidentified interstellar line at 1270.96 Å is assigned to the strongest line of the 4d-X 2Π transition of CH.

Subject headings: ISM: lines and bands — ISM: molecules — methods: laboratory — molecular data — ultraviolet: ISM

1. INTRODUCTION

The visible A 2Σ–X 2Π and near-ultraviolet B 2Σ–X 2Π and C 2Σ+–X 2Π bands of the CH molecule were among the first molecular spectra to be identified in astronomical sources. The latter encompass diffuse interstellar clouds (Swings & Rosenfeld 1937; McKellar 1940a, 1940b), comets (Nicolet 1938; Dufay 1940), and stellar atmospheres, including that of the Sun (Stenstine, Svensson, & Olsen 1938; Wildt 1941; Babcock 1945). Later, CH in dark interstellar clouds was detected by radioastronomy using A-doubling transitions of the X 2Π ground state (Rydebeck, Eldric, & Irvine 1973; Turner & Zuckerman 1974). Since then, many studies have been made of this widespread molecule.

A great interest exists in the electronic spectrum of CH in the vacuum ultraviolet region. In 1969, Herzberg & Johns reported a number of bands of the CH and CD molecules observed in absorption in the laboratory following the flash photolysis of diazomethane, H2CNN (Herzberg & Johns 1969). Among these is a Rydberg series assigned as nd-X 2Π, of which the first member, n = 3 near 1370 Å, shows rather irregular rotational structure. The upper states of this band were referred to as G(2Σ, 2Π, 3Σ) by Herzberg & Johns (1969), but here the simpler designation 3d will be used. About 45 lines were listed but not assigned by these authors. The corresponding band of the isotope CH3 was also observed.

Probably the main point of astronomical interest in this spectrum is the existence of close wavelength coincidences of lines with three unidentified (UID) interstellar lines: UID1 at 1368.74 Å (73059.9 cm⁻¹), UID2 at 1369.13 Å (73039.1 cm⁻¹), and UID3 at 1370.87 Å (72946.4 cm⁻¹; Cardelli, Savage, & Ebbets 1971; Savage, Cardelli, & Sofia 1992), in the spectrum of the diffuse interstellar clouds toward the O-type star ζ Ophuchi. The corresponding laboratory wavenumbers listed by Herzberg & Johns (1969) are 73059.6, 73039.4, and 72946.2 cm⁻¹. The agreement seems adequate, given that the laboratory spectrum has an apparent resolution of about 1.3 cm⁻¹ and that at laboratory temperatures several transitions probably contribute to each “line.” The interstellar UID2 line is broader than the others and is classified as the only known diffuse interstellar band (DIB) in this wavelength region (Tripp, Cardelli, & Savage 1994; Morton & Noreau 1994; Gacinski, Sikorski, & Kaczmarczyk 1997). The corresponding laboratory line is also noticeably broader than the others, but it is partially obscured by a neighboring stronger line. It is therefore difficult to say whether the profile of the laboratory line agrees with the asymmetry claimed for the observed line in ζ Oph (Tripp et al. 1994), which may not in any case be an intrinsic property of this line because the profile toward the star X Per (see data set 22c00101h of the Hubble Space Telescope archive) seems to be asymmetric in the opposite sense.

The equivalent widths of the three UID lines toward ζ Oph, 3.80, 5.89, and 0.84 mÅ, respectively (Tripp et al. 1994), are comparable to those for the visible and near-ultraviolet lines for which Lien (1984) gives values of 17.8 for (A–X); 5.0, 3.3, and 1.6 for (B–X); and 7.4, 5.0, and 4.0 for (C–X) mÅ. In their qualitative descriptions, Herzberg & Johns (1969) imply that the intensity of the 3d–X band of CH is similar to those of the above three band systems; in fact, they say, “The expected lines of this system... would be the strongest interstellar lines of CH (Herzberg & Johns 1969).” The precise basis for this statement is not clear. For an optically thin line, the square of the transition moment is proportional to W_d/λ, where W_d is the equivalent width, and toward ζ Oph this quantity has the value 2.8 × 10⁻⁶, 4.3 × 10⁻⁶, and 0.6 × 10⁻⁶ for the three UID lines from the data of Tripp et al. (1994), compared to 4.1 × 10⁻⁶ for the strongest assigned line, the A 2Σ–X 2Π line near 4300 Å (Danks, Federman, & Lambert 1984).

Thus, there is a prima facie case that the three UID lines may be attributed to CH, but a definite identification requires a rotational assignment of the laboratory spectrum. The temperature of the diffuse clouds toward ζ Oph is sufficiently low that only the lowest rotational levels of CH are populated (Herbig 1968; Lien 1984). These are the e and f A-doubling components of the J = 0.5 level of the 2Π₁/₂ state, which are separated in energy by 0.11 cm⁻¹. Transitions between these components are responsible for the radio-frequency spectrum near 3300 MHz (Rydebeck et al. 1973, 1974; Turner & Zuckerman 1974). Thus, it is necessary to know whether the above lines in the laboratory spectrum correspond to the strongest transitions from the
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The rotational energy levels of the \( X^2 \Pi \) ground electronic state of CH are well known. Here the effective Hamiltonian and constants of Zachwieja (1995) are used. His work incorporates the results of new measurements of the \( A^2 \Pi - X^2 \Pi \) band system, as well as of a number of recent high-resolution studies. For the \( 3d \) of Rydberg complex, the computer program employed here is one that has been applied recently to Rydberg-Rydberg transitions of ArI and ArR (Dabrowski et al. 1996, 1998a, 1998b; Nowlan, Tokaryk, & Watson 2001). It is applicable to a diatomic molecule with an ionic core in a \( ^2 \Sigma^+ \) electronic state, in this case the ground state of CH\(^+\). Because of the rather low resolution, the only terms of the effective Hamiltonian employed here are

\[
H = T_0 + G_0^{(2)}(P_0 - \frac{3}{2}P) + G_0^{(4)} \times \left[ \frac{1}{2}P^2 - 1 - \frac{3}{2}P(P - 2) + \frac{3}{4}P(P - 2) \right] + B_0 R^2 - D_0 R^4,
\]

where

\[
R \equiv J^+ \equiv N^+ = J - I - s
\]

is the total angular momentum without nuclear spin of the CH\(^+\) core. \( J \) is the total angular momentum without nuclear spin of the neutral CH molecule, and \( I \) and \( s \) are the orbital and spin angular momenta of the Rydberg electron. In equation (1), the operators with coefficients \( G_0^{(2)} \) and \( G_0^{(4)} \) are unnormalized second- and fourth-rank spherical tensor components of the types \( T(2, 0) \) and \( T(4, 0) \), respectively (Zac 1988). For a \( d \) complex, a choice of \( T_0, G_0^{(2)} \), and \( G_0^{(4)} \) is equivalent to choosing independent term values for the \( 2^2 \Sigma^+, 2^2 \Pi \), and \( 2^2 \Delta \) components. The intensities in the spectra calculated here are based on pure \( \delta \alpha \) transition moments between the basis functions (Dabrowski et al. 1996).

Initially, attempts were made to assign the laboratory spectrum without reference to the interstellar lines, but this proved to be difficult because the rotational structure is rather complex, and at least four parameters \([T_0, G_0^{(2)}, G_0^{(4)}, \text{and } B_0]\) have to be adjusted. On the other hand, for the interstellar lines only low rotational levels are involved, and it is sufficient to take \( B_0 \) equal to the value for the \( X^2 \Sigma^+ \) state of the CH\(^+\) core (Huber & Herzberg 1979). The \( D_0 \) value was held throughout at the value for CH\(^+\).

On the assumption that the UID lines are \( R(0.5) \) transitions, from the \( X^2 \Pi \)-state term value table of Zachwieja (1995) one expects \( Q(1.5) \) transitions with the same upper levels at approximately 17.8 and 67.0 cm\(^{-1}\) lower in wave-number in the laboratory spectrum. (The precise values depend on which A-doubling components are involved.)
There are also $P(2.5)$ transitions with the same upper levels, but these are calculated to be much weaker. The presence of the $Q(1.5)$ lines is borne out by the wavenumber list of Herzberg & Johns (1969), with some additional information from the scanner spectra. There are lines at 17.7 and 66.7 below 73059.6 lines at 18.2 and 67.1 (sh) below 73039.4, and lines at 17.9 and 66.8 (sh) below 72946.2 (all in cm$^{-1}$); the (sh) lines are shoulders seen in the scanner spectrum. These combination-difference features therefore support the proposed identification. By adjusting the parameters to $T_0 = 72995.2$, $G_0^G = 21.6$, and $G_0^Q = -39.2$ (all in cm$^{-1}$), it was found that these lines could be fitted and gave approximately the correct relative intensities as the strongest three lines of the band at low temperature (arbitrarily taken as 10 K).

The spectrum calculated at a temperature of 300 K with these parameters immediately reproduced the general features of the laboratory spectrum, particularly the $Q$-form "hump" at 73021 cm$^{-1}$ and the general distribution of the lines. It became possible to assign lines of higher J and to adjust $B_0$ of the 3d complex to improve the fit.

Eventually, the four parameters $T_0$, $G_0^G$, $G_0^Q$, and $B_0$ of the 3d complex were fitted to 31 lines with a standard deviation of 0.21 cm$^{-1}$. The assignments of the fitted lines are given in Table 1. Adjustment of other parameters did not improve the fit significantly. There are a number of additional lines, but in general they were rejected when included in the fit. This may be owing to perturbations by vibrational levels of other electronic states, which may be relatively dense at this energy. The final parameters are then $T_0(3d) = 72995.18(7)$, $G_0^G(3d) = 32.54(4)$, $G_0^Q(3d) = 36.68(7)$, $B_0(3d) = 13.400(3)$, and $D_0(3d) = 0.0014(6)$, all in cm$^{-1}$ with standard deviations in units of the last quoted decimal place in parentheses. The resulting term values of the three electronic components obtained by putting $R = 0$ in equation (1) are

\[ T_0(3d^2 \Sigma - 2^\Pi) = 72874.78, \quad T_0(3d^2 \Sigma - 2^\Pi) = 73022.91, \quad T_0(3d^2 \Delta) = 73027.90 \text{ cm}^{-1}. \]

Trace a of Figure 1 presents the calculated unBroadened spectrum at 10 K, showing that the three strongest lines agree with the observed UID lines. Clearly, additional lines might be observable with higher sensitivity. The calculated wavenumbers of the lines with $J^* = 0.5$ are given in Table 2. There are two additional weak lines at 72992.3 and 73021.3 cm$^{-1}$ in trace a of Figure 1 that originate from $J^* = 1.5$ levels. Trace b of Figure 1 shows the calculated spectrum at 300 K, broadened with a Lorentzian profile, with FWHM of 1.3 cm$^{-1}$, approximately that of the sharpest line in the observed spectrum. Trace c is described in §4, and trace d is a scan of the observed laboratory spectrum. The fit of the line positions is good near the band center but degrades toward the wings. The extra intensity of the observed spectrum in the range 72900–73000 cm$^{-1}$ may possibly be a result of deviations from the simple transition moments assumed in the calculations.

From the above parameters of CH, we can calculate an approximate spectrum of CD. The parameters $T_0$, $G_0^G$, and $G_0^Q$ of the 3d complex should be approximately unchanged, apart from the unknown effects of zero-point vibrations. Parameters $B_0$ and $D_0$ for CD are calculated on the assumption that the ratios $B_0(\text{CD})/B_0(\text{CH})$ and $D_0(\text{CD})/D_0(\text{CH})$ for the 3d complex are the same as for the X state; this procedure allows partially for the effects of the zero-point vibration on the rotational structure. For the X state of CD, the parameters of Brown & Ewenson (1989) are employed. The resulting spectrum at 300 K with Lorentzian broadening with FWHM of 1 cm$^{-1}$, is shown in Figure 2, along with a trace of the laboratory spectrum (Herzberg & Johns 1969) for comparison. To improve the agreement, the calculated spectrum has been shifted to lower wavenumbers by 30 cm$^{-1}$, corresponding to the isotopic shift of the 0-0 vibrational band. It is seen that the general distribution of lines in this band is well reproduced, although it might be difficult to make specific line assignments because of the congestion in the laboratory spectrum. This good qualitative agreement for CD lends support to the values of the parameters obtained for the CH isotope.
TABLE 2

<table>
<thead>
<tr>
<th>( R_{\text{cld}} )</th>
<th>( N^a )</th>
<th>( c/d^a )</th>
<th>( F^a )</th>
<th>( F''^a )</th>
<th>( \delta^a )</th>
<th>( \iota^a )</th>
<th>Observed</th>
<th>Width (^b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.582</td>
<td>1</td>
<td>c</td>
<td>( F_{\lambda}(0.5) )</td>
<td>( F_{\lambda}(0.5) )</td>
<td>72938.87</td>
<td>1.232</td>
<td>Narrow</td>
<td></td>
</tr>
<tr>
<td>1.582</td>
<td>1</td>
<td>c</td>
<td>( F_{\lambda}(1.5) )</td>
<td>( F_{\lambda}(0.5) )</td>
<td>72938.87</td>
<td>0.616</td>
<td>Narrow</td>
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</tr>
<tr>
<td>2.000</td>
<td>0</td>
<td>c</td>
<td>( F_{\lambda}(0.5) )</td>
<td>( F_{\lambda}(0.5) )</td>
<td>72940.67</td>
<td>0.632</td>
<td>Narrow</td>
<td></td>
</tr>
<tr>
<td>1.191</td>
<td>2</td>
<td>d</td>
<td>( F_{\lambda}(1.5) )</td>
<td>( F_{\lambda}(1.5) )</td>
<td>72946.14</td>
<td>2.708</td>
<td>UID3</td>
<td>Narrow</td>
</tr>
<tr>
<td>1.001</td>
<td>2</td>
<td>d</td>
<td>( F_{\lambda}(1.5) )</td>
<td>( F_{\lambda}(1.5) )</td>
<td>73037.92</td>
<td>7.022</td>
<td>UID2</td>
<td>Broad</td>
</tr>
<tr>
<td>1.768</td>
<td>2</td>
<td>d</td>
<td>( F_{\lambda}(1.5) )</td>
<td>( F_{\lambda}(1.5) )</td>
<td>73059.93</td>
<td>4.600</td>
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<td>Narrow</td>
</tr>
<tr>
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<td>1</td>
<td>d</td>
<td>( F_{\lambda}(0.5) )</td>
<td>( F_{\lambda}(0.5) )</td>
<td>73088.87</td>
<td>0.467</td>
<td>Broad</td>
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<tr>
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<td>1</td>
<td>d</td>
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<td>( F_{\lambda}(0.5) )</td>
<td>73088.87</td>
<td>0.933</td>
<td>Broad</td>
<td></td>
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<tr>
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<td>1</td>
<td>d</td>
<td>( F_{\lambda}(0.5) )</td>
<td>( F_{\lambda}(0.5) )</td>
<td>73117.58</td>
<td>0.325</td>
<td>Narrow</td>
<td></td>
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<tr>
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<td>( F_{\lambda}(0.5) )</td>
<td>73117.58</td>
<td>0.162</td>
<td>Narrow</td>
<td></td>
</tr>
</tbody>
</table>

Note.—Notation as in Table 1.

\(^a\) Relative intensity, assuming pure \( d \rightarrow \iota \) transition moments.

\(^b\) Since both sets of lines are probably predissociated, the descriptions "narrow" and "broad" are only relative terms.

are probably an indication of the difficulty of calculating Rydberg states using limited basis sets, particularly for a molecule like CH that has a number of lower lying electronic states. The splittings between the 3d components found here are of a similar order of magnitude to those observed in the 3d states of BeI (Cleraux & Colin 1991) and BH (Johns & Lepard 1975), which were fitted with \( G_3^2(3d) = 2 \lambda_3 = 82.6 \) and 47.4 cm\(^{-1}\), respectively, assuming \( G_3^2(3d) = 0 \) in both cases.

The column density of CH toward \( \zeta \) Oph is approximately \( N(CH) = 10^{13.30} \) cm\(^{-2}\) (Federman & Lumbert 1988). The sum of the equivalent widths of the three 3d-X U1D lines (Trapp et al. 1994) implies an oscillator strength of \( f = 0.025 \) plus small contributions from unobserved lines for the 3d-X \( ^2\Pi \) transition of CH. From the calculated relative intensities in Table 2, an estimate of \( f = 0.033 \) is obtained for the whole band. This agrees somewhat better with the ab initio value of 0.0362 of Barsuhn & Nesbitt (1978) than with van Dishoeck's (1987) value of 0.069. The relative oscillator strengths of 1:3:6 for the \( ^2\Sigma^+ \), \( ^2\Pi \), and \( ^2\Delta \) components are assumed here, whereas Barsuhn & Nesbitt (1987) calculated absolute values of 0.0012, 0.0015, and 0.0036, and van Dishoeck (1987) calculated values of 0.005, 0.030, and 0.034. The comparison in Figure 1 shows that more intensity is required in the calculated spectrum near 72950 cm\(^{-1}\), but it would be best to allow for the effects of varying line widths before adjusting the relative transition moments.

4. LINE WIDTHS

The lines UID1, UID2, and UID3 have FWHMs of 1.2, 2.6, and 0.8 cm\(^{-1}\), respectively, toward \( \zeta \) Oph (Trapp et al. 1994). The velocity structure of CH in this line of sight is somewhat complex but gives an FWHM of approximately 2 km s\(^{-1}\) for a single A-doublet component, from the work of Crawford et al. (1994). This translates to 0.5 cm\(^{-1}\) near 1370 Å. Thus, it seems that the three levels are broadened beyond the velocity width, and the most likely explanation is predissociation.

The upper levels of the U1D lines are assigned here as \( J = 1.5 \) levels of parity types \( f, e, \) and \( f \), respectively, which correspond to absolute parities of +, −, and +. These levels all have values of \( N = 2 \) for the total angular momentum without spin. Levels with parities \( \pm (1)^2 \) and \( \mp (1)^2 \) are described as \( c \) and \( d \) levels, respectively (Herzberg 1989; Mizushima 1975, p. 86; Watson 1991); so the upper levels are of types \( c, d, \) and \( e \), respectively. If only interactions that obey the selection rules \( \Delta S = 0 \) (Herzberg 1989) are considered, then dissociative states with \( \Lambda \neq 0 \) will predissociate both \( c \) and \( d \) levels, but a \( \Delta \) state will predissociate either only the \( c \) levels if it is \( ^2\Sigma^+ \) or only the \( d \) levels if it is \( ^2\Sigma^- \) (Herzberg 1989). An example for a Rydberg complex of the NO molecule has been described by Huber et al. (1987).

From these considerations, the widths of UID1 and UID3 beyond the velocity width may be owing to predissociation by either \( ^2\Sigma^+ \) or \( ^2\Sigma^- \), but the extra width of UID2 can be attributed to predissociation by \( ^2\Pi \) states. The 3d complex is some 46000 cm\(^{-1}\) above the dissociation limit of the known \( P^2\Sigma^- \) state (Herzberg & Johns 1969). The \( ^2\Sigma^- \) state is calculated to be in the same energy region as the 3d complex (Barsuhn & Nesbitt 1978; van Dishoeck 1987) and may also be involved in this predissociation. The predissociation must be heterogeneous and \( N \) dependent because the three components of the 3d complex have different vibronic symmetry from a \( ^2\Sigma^- \) state. Without more detailed knowledge, it is not possible to model the \( N \) dependence of this predissociation.

The apparent resolution of the laboratory spectrum is comparable to the widths of UID1 and UID3, but the extra width of the line corresponding to UID2 is noticeable, although it is difficult to quantify because of the overlapping strong line. One feature of the observed laboratory spectrum that can be attributed to this predissociation is the unresolved nature of the hump near 73021 cm\(^{-1}\). All of the calculated levels of significant intensity in the range 72995-73040 cm\(^{-1}\) have upper levels of \( d \) parity and are therefore subject to the extra predissociation. To illustrate the effect of differential broadening of the \( c \) and \( d \) upper levels, trace \( c \) of Figure 1 is a calculated spectrum with Lorentz profiles with FWHM of 1.3 cm\(^{-1}\) for lines with \( c \) upper levels and 2.6 cm\(^{-1}\) from the width of the interstellar UID2) for \( d \) upper levels. This largely simulates the shape of the 73021 cm\(^{-1}\) "hump." However, it is an oversimplification to assume that the broadening is the same for all \( d \) upper levels. A discussion of the various predissociation processes for CH in interstellar clouds, together with earlier references, is given by van Dishoeck (1987).

5. THE 4d-X TRANSITION

The 1370 Å laboratory band is the first member of a Rydberg series that is shown in Figure 5 of the paper by Herzberg & Johns (1969). The second member, 4d-X, is also
of significant intensity, and it seemed of interest to calculate its rotational structure at low temperatures. Using the ionization potential of 85.90 cm\(^{-1}\) (Herzberg & Johns 1969), the 3d complex has effective principal quantum number \(n^* = 2.922\). The 4d complex is then predicted to have \(n^* = 3.922\), which gives \(T_0 = 78715, G_{0}^{d} = 9.34, G_{0}^{d} = 15.16, B_{0} = 13.7120,\) and \(D_{0} = 0.0014\) cm\(^{-1}\), where the \(n^*\) \(\pm 3\) selecting law (Berg et al. 1993; Jakubek & Field 1997) is applied to \(G_{0}^{d}, G_{0}^{d},\) and \(|B_{0}(\text{CH})| = 0.6\) CH \(^{-1}\).

A 4d-X spectrum calculated at 300 K with these parameters gives a strong Q-form peak at 1270.3 Å. To match the observed peak at 1271.3 Å (Herzberg & Johns 1969), it is necessary to decrease \(T_{0}(4d)\) to 78653 cm\(^{-1}\). Then, the spectrum calculated at 10 K shows two strong lines at 78633.4 and 78679.2 cm\(^{-1}\), corresponding to \(\lambda \lambda 1271.7\) and 1271.0, as well as a number of weaker lines, as shown in Figure 3.

The strongest line agrees well in wavelength with a UID line in \(\xi\) Oph reported at 1270.96 Å by Federman et al. (1995); there may also be a very weak feature near 1271.7 Å close to noise level in their spectrum. The equivalent width of 0.75 mA reported for the UID line (Federman et al. 1995) appears qualitatively consistent with the relative intensities of the 4d-X and 3d-X bands in the laboratory spectrum (Herzberg & Johns 1969). Thus, it is proposed that the \(\lambda 1270.96\) UID line is also attributable to the CH molecule. Unfortunately, the original plate of the published spectrum (Herzberg & Johns 1969) has not been located, so it is not possible to test the present prediction of the rotational structure in more detail.

6. SUMMARY

This paper presents strong evidence that the UID1, UID2, and UID3 lines of Cardelli et al. (1991) and the UID line at 1270.96 Å of Federman et al. (1995) are attributable to interstellar CH. For the first three lines, this interpretation is consistent with a rotational assignment of the laboratory spectrum of Herzberg & Johns (1969). More detailed analysis of higher resolution laboratory spectra is desirable but may be difficult because of the predissociation widths of the lines. Probably the most valuable new data would be laboratory spectra with lower rotational temperatures.

The fourth UID line is the strongest line of the 4d-X band at low temperatures, based on using the expected \(n^*\) \(\pm 3\) dependence of the molecular parameters from the 3d-X band, together with an adjustment to fit the feature observed in the laboratory spectrum. High-resolution laboratory spectra of this band would clearly be desirable.

I am grateful to Dr. Donald C. Morton for bringing this problem to my attention and for informing me of the paper of Federman et al. (1995), to Dr. Dennis W. Tokarz for using the optical scanner technique to produce the scans of the observed spectra reproduced in Figures 1 and 2, and to Dr. Zygmun Jakubek for an important suggestion.

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