High-resolution Fourier-transform emission spectroscopy of the $A^1\Pi$–$X^1\Sigma^+$ system of AlH

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The emission spectrum of the $A^1\Pi$–$X^1\Sigma^+$ system of AlH, excited in a hollow-cathode discharge lamp, has been observed at high resolution with a Fourier-transform spectrometer. The rotational lines in the 0–0 and the 1–1 bands have been measured with a precision of ± 0.001 cm^{-1}. The present measurements provide a considerable improvement over the previous data of Zeeman and Ritter [Can. J. Phys. **32,** 555 (1954)]. The present data, combined with the previous high-resolution measurements of the 1–0 vibration–rotation band by White et al. [J. Chem. Phys. **99,** 8371 (1993)] and the $J = 1$–0 pure rotational line of Goto and Saito [Astrophys. J. **452,** L147 (1995)] have been used to determine improved molecular constants for the $A^1\Pi$ state. © 1996 Optical Society of America

1. Introduction

In the past decade, there has been growing interest in experimental and theoretical studies of the AlH molecule because of its importance in several areas of science, including astrophysics[1–4] and chemistry.[5–7] AlH has been detected in the spectra of M-, S-, Sp-, and C-type stars[1–3] and in sunspots.[4] Erlandson and Cool[5] have explored the possibility of using AlH in a dissociation laser operating in the visible region. AlH is frequently produced as a photodissociation product of Al-containing organometallic precursors in chemical-vapor-deposition experiments.[6] AlH is also a candidate for an advanced chemical rocket propellant that is synthesized when metal-hydride molecules are trapped in cryogenic H_2 matrices.[7]

The spectrum of AlH has been known since the beginning of this century,[8] when it was initially described as being due to a compound of aluminum and hydrogen. Since then, the visible spectra of this molecule have been investigated by several workers,[9–15] and a strong band with a Q-head at 4259 Å has been assigned as the 0–0 band of the $A^1\Pi$–$X^1\Sigma^+$ transition.[12,13] The most complete analysis of this transition is provided by Zeeman and Ritter,[15] who studied nine bands involving the vibrational

levels $v = 0$, 1 in the $A^1\Pi$ state and $v = 0$–4 in the ground $X^1\Sigma^+$ state. These studies indicated that the $A^1\Pi$ state is involved in predissociation. The bands involving the $v = 0$, 1 vibrational levels of the $A^1\Pi$ state show a sharp cutoff in the rotational structure because of predissociation. In addition to the $A^1\Pi$ state, several other higher singlet and triplet excited states have been observed in emission as well as in absorption.[16]

AlH has also been the subject of many theoretical studies.[17–26] The properties of the ground and low-lying excited states have been predicted, most notably by Meyer and Rosmus,[17] Matos et al.,[22] and Bauschlicher and Langhoff.[23] Matos et al.,[22] have reported the molecular constants and potentials for the $X^1\Sigma^+$, $A^1\Pi$, and $C^1\Sigma^+$ states, and Bauschlicher and Langhoff[23] have calculated the dipole moments as well as the lifetimes. The spectroscopic properties of the Rydberg states were calculated by Grimaldi et al.,[26] and the rotational g values were predicted by Sauer and Ogilvie.[27] The Franck–Condon factors for the $A^1\Pi$–$X^1\Sigma^+$ transition were calculated from Rydberg–Klein–Rees (RKR) potentials by Huron.[28]

The vibration–rotation spectra of AlH and AlD have also been studied by several groups.[29–33] The most complete analysis of the AlH and AlD infrared bands has been provided by White et al.,[32] who measured several vibration–rotation bands of AlH (from 1–0 to 5–4) and AlD (from 1–0 to 7–6) with a precision of ± 0.0002 cm^{-1}. This work provided the most accurate ground-state molecular constants for AlH and AlD.

The lifetimes of 66 ± 4 ns for $v = 0$ and 83 ± 6 ns for $v = 1$ have been measured by Baltayan and

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Nedelec[34] for the $A^1\Pi$ state by laser-induced fluorescence, and Rice *et al.*[35] have determined vibrational band strengths. Zhu *et al.*[36] have also measured the $A^1\Pi$–$X^1\Sigma^+$, $b^3\Sigma^-$–$X^1\Sigma^+$, and $C^1\Sigma^+$–$X^1\Sigma^+$ line positions by laser-induced fluorescence. In other laser experiments, the pulsed-laser vaporization technique has been used to react Al atoms with H_2 in solid Kr matrices by Parnis and Ozin[37] and in Ar by Chertihin and Andrews[38] in order to detect the matrix infrared spectra of AlH, AlH_2, AlH_3, and other species. Hwang and Dagdigian[39] observed the 0–0 band of the $A^1\Pi$–$X^1\Sigma^+$ system of AlH during the search for the electronic spectra of the KrAlH van der Waals complex in a free jet expansion. They tried to use the previously reported AlH line positions[15,40] of the 0–0 band of the $A^1\Pi$–$X^1\Sigma^+$ system for calibrating their laser excitation spectra but encountered problems. They noticed that there are large discrepancies between the published line positions[40] and those calculated from the constants of Zeeman and Ritter.[15] The recent laser measurements[36] of the $A^1\Pi$–$X^1\Sigma^+$ transition differ from both of the older measurements.[15,40] Dagdigian and the astronomer D. Carbon brought these problems with the $A^1\Pi$–$X^1\Sigma^+$ transition of AlH to our attention. We had inadvertently recorded spectra of the 0–0 and 1–1 bands of the $A^1\Pi$–$X^1\Sigma^+$ AlH transition some years ago. Our analysis of this archival data provided greatly improved line positions and molecular constants, which may prove useful in the monitoring of AlH in both laboratory and extraterrestrial environments.

2. Experimental

The spectrum of AlH was observed by accident during a search for the AlC molecule by the use of a hollow-cathode lamp. The cathode was prepared when a mixture of Cu and Al_4C_3 powders (3:1 ratio) was pressed in a hole in a Cu block. The block was then bored through to provide a layer of Cu/Al_4C_3 on the inside walls of the hollow cathode. The lamp was operated at 470 V and 400-mA current with a slow and steady flow of ~1.5 Torr of Ar gas. No H_2 was added but evidently there were enough H-containing impurities to provide a strong AlH spectrum.

The spectra were recorded with the 1-m Fourier-transform spectrometer associated with the McMath–Pierce Solar Telescope of the National Solar Observatory. The spectra in the 17,000–25,000-cm^{-1} region were recorded with GaAs photomultiplier detectors and 400-nm red-pass and 550-nm blue-pass filters. In total, 100 scans were coadded in ~4 h of integration at a resolution of 0.05 cm^{-1}.

The spectral line positions were extracted from the observed spectra by the use of a data-reduction program called PC-DECOMP developed by J. Brault. The peak positions were determined by the fitting of a Voigt line-shape function to each spectral feature.

In addition to the AlH bands, the final spectra also contained Al and Ar atomic lines as well as AlO and
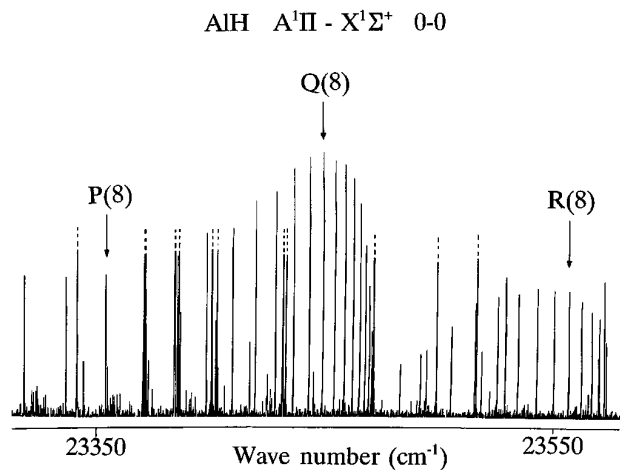

Fig. 1. Compressed portion of the spectrum of the 0–0 band of the $A^1\Pi$–$X^1\Sigma^+$ system of AlH.

CH molecular bands. The spectra were calibrated with the measurements of Ar atomic lines made by Norlén.[41] The AlH lines have widths of ~0.1 cm^{-1} and appear with a maximum signal-to-noise ratio of ~60:1 in the 0–0 band so the absolute accuracy and precision of the measurements are expected to be of the order of ±0.001 cm^{-1}. The 1–1 band is much weaker than the 0–0 band, so the precision of measurements in this band ranges from ±0.002 to ±0.003 cm^{-1}.

3. Results and Discussion

Each band of the $A^1\Pi$–$X^1\Sigma^+$ transition consists of a single R-, a single P-, and a single Q-branch, as expected for a $^1\Pi$–$^1\Sigma^+$ transition. A compressed part of the spectrum of this transition has been provided in Fig. 1. The R-branch lines are slightly weaker than the P-branch lines, and the Q-branch is the most intense. The lines in the 1–1 band are less than 50% of the intensity of the 0–0 band. The rotational lines could be followed up to $R(16)$, $Q(17)$,
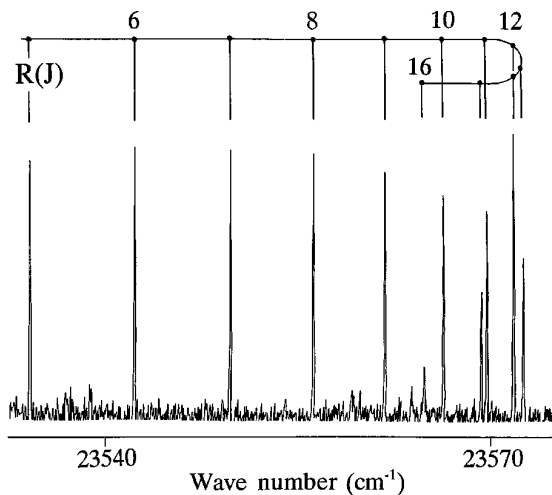

Fig. 2. Expanded portion of the R-branch of the 0–0 band near the head that shows the breaking off in intensity at $R(16)$.

and $P(18)$ in the 0–0 band and up to $R(6)$, $Q(7)$, and $P(8)$ in the 1–1 band. No rotational perturbations were observed in these two bands. As reported in the previous studies that used low-pressure sources, a sharp cutoff in the intensity is observed. In the present spectrum the lines could not be extended beyond $J' = 17$ in the 0–0 band (see Fig. 2). The intensity of the lines with $J' = 17$ is much smaller than the lower J members in the branches, indicating the presence of predissociation for $J' \geq 17$. The same phenomenon is observed in the 1–1 band for $J' \geq 7$.

To determine the rotational constants, the observed line positions were fitted with the following customary energy-level expressions for $^1\Sigma^+$ [Eq. (1)] and $^1\Pi$ [Eq. (2)] states:

$$F_v(J) = T_v + B_vJ(J+1) - D_v[J(J+1)]^2 + H_v[J(J+1)]^3$$
$$+ L_v[J(J+1)]^4, \qquad (1)$$

$$F_v(J) = T_v + B_vJ(J+1) - D_v[J(J+1)]^2 + H_v[J(J+1)]^3$$
$$+ L_v[J(J+1)]^4 \pm 1/2\{q_vJ(J+1)$$
$$+ qD_v[J(J+1)]^2\} \qquad (2)$$

with the plus sign for e-levels and the minus sign for

f-levels in the Λ-doubling terms. The measurements of the 1–0 vibration–rotation band by White *et al.*[32] as well as the $J = 1$–0 hyperfine-corrected pure rotational line [377738.266(40) MHz] recently measured by Goto and Saito[42] were also incorporated into this fit. The infrared transitions and the measurements obtained in the present work were given appropriate weights based on resolution, signal-to-noise ratio, and extent of blending. We observe that the line positions reproduced in Herzberg's book[40] differ from the present measurements by substantial amounts ranging from +0.03 to +0.20 cm^{-1}. In particular, the $R(0)$ line of the 0–0 band is off by 0.60 cm^{-1}. Our measurements also differ from those of Zhu *et al.*[36] by ~0.4 cm^{-1}. The wave numbers of the observed rotational lines in the 0–0 and the 1–1 bands are provided in Table 1, and the constants obtained from the final fit are provided in Table 2. Interestingly, the constants of Zeeman and Ritter[15] turn out to have been relatively reliable, although they were determined by graphical methods rather than by least-squares fits. For example, their 0–0 origin[15] of 23470.93 cm^{-1} compares with our value (Table 2) of 23470.90129 cm^{-1} and their $B_0' = 6.0207$ cm^{-1} compares with our value of 6.020328 cm^{-1}.

The breaking off in the intensity of the branches

Table 1. Vacuum Wave Numbers (cm^{-1}) of the $A^1\Pi$–$X^1\Sigma^+$ System of AlH^a

J	$R(J)$	O–C	$Q(J)$	O–C	$P(J)$	O–C
			0–0 Band			
0	23482.9489	0.0012				
1	23494.4284	0.0030	23470.3306	−0.0005		
2	23505.3151	0.0027	23469.1828	−0.0016	23445.1540	−0.0025
3	23515.5797	−0.0009	23467.4493	0.0012	a	
4	23525.1943	−0.0001	23465.1047	0.0016	23417.2333	−0.0018
5	23534.1100	−0.0018	23462.1235	0.0005	23402.4427	−0.0006
6	23542.2801	−0.0028	23458.4745	−0.0001	23387.0759	0.0001
7	23549.6490	−0.0007	23454.1179	0.0007	a	
8	23556.1458	0.0002	23449.0011	−0.0008	23354.5075	0.0013
9	23561.6945	0.0004	23443.0708	−0.0005	23337.2287	−0.0018
10	23566.2069	−0.0005	23436.2583	0.0004	23319.2314	0.0007
11	23569.5856	0.0006	23428.4834	0.0004	23300.4473	0.0003
12	23571.7111	−0.0002	23419.6553	−0.0001	23280.8100	0.0019
13	23572.4509	−0.0026	23409.6681	−0.0002	a	
14	23571.6555	−0.0022	23398.3971	−0.0009	23238.6123	0.0000
15	23569.1467	0.0015	23385.7016	0.0021	23215.8391	0.0002
16	23564.7077	−0.0006	a		23191.7705	−0.0004
17			23355.3102	0.0002	23166.2453	0.0003
18					23139.0683	−0.0002
			1–1 Band			
0	22939.1958	0.0085 b				
1	22948.0871	0.0030	22926.9421	0.0013		
2	22955.2535	0.0023	22923.5846	−0.0017	22902.4933	−0.0032
3	22960.6181	−0.0016	a		22886.9538	−0.0075 b
4	22964.0923	−0.0047	22911.6373	−0.0026	a	
5	22965.5625	−0.0009	22902.9112	0.0079 b	22850.7825	0.0031
6	22964.8685	0.0000	22892.1869	0.0016	22830.0022	−0.0029
7			22879.3428	−0.0013	22807.3162	0.0021
8					22782.5897	0.0169 b

^{a}a indicates overlapping by atomic lines and b indicates blending.

Table 2. Spectroscopic Constants (cm⁻¹) for the $A^1\Pi$–$X^1\Sigma^+$ System of AlH

Constants	$X^1\Sigma^+$		$A^1\Pi$	
	$v = 0^a$	$v = 1$	$v = 0$	$v = 1$
T_v	0.0	1625.069602(31)	23470.90129(49)	24553.6781(15)
B_v	6.30072554(90)	6.1172960(10)	6.020328(27)	5.28835(27)
$10^3 \times D_v$	0.3653934(74)	0.3589520(80)	0.63312(39)	1.145(11)
$10^6 \times H_v$	0.015110(21)	0.014916(20)	−0.0586(20)	−1.08(13)
$10^{10} \times L_v$	−0.00816(18)	−0.00793(16)	−1.713(33)	—
$10^3 \times q_v$	—	—	8.3212(70)	6.664(38)
$10^6 \times q_{D_v}$	—	—	−3.398(30)	—

^{a}Numbers in parentheses are one standard deviation in the last digit.

with $J' \geq 17$ in the 0–0 band and with $J' \geq 7$ in the 1–1 band is consistent with the previous observations in the emission spectra from low-temperature arc or discharge sources. Zeeman and Ritter[15] observed broad rotational lines beyond the breaking-off point. This was due to the use of a King furnace at high temperature (~2100 °C) and high pressure (40 to 60 Torr) to establish thermal equilibrium. In this case the population of the $A^1\Pi$ rotational levels, even those affected by predissociation, are determined simply by the temperature.[40] The sharp cutoff in the intensity of the rotational lines is due to predissociation by rotation, as has been discussed in detail by Herzberg.[40]

The inclusion of the 1–0 vibration–rotation measurements of White et al.[32] and the $J = 1$–0 pure rotational line of Goto and Saito[42] helps to determine a precise set of molecular constants for the ground and the excited states. The rotational constants for the $A^1\Pi$ state have been used to evaluate the equilibrium rotational constants provided in Table 3. The present constants agree well with those of Zeeman and Ritter[15] within the quoted standard deviation. The new excited-state constants, however, are expected to be more precise than the values of Zeeman and Ritter.[15] The B_e value of 6.38632(14) cm⁻¹ for the $A^1\Pi$ state provides the equilibrium bond length of 1.648328(17) Å, similar to the value of 1.6453622(21) Å in the ground state.[32] The $A^1\Pi$–$X^1\Sigma^+$ transition is very diagonal,[28] and we did not observe any off-diagonal bands in our spectra because of their weak intensity. This observation is consistent with the Franck–Condon calculations of the A–X system by Huron[28] and the Einstein A coefficients (15.2 × 10⁶

s⁻¹ and 10.2 × 10⁶ s⁻¹ for 0–0 and 1–1 bands) of this transition, as reported by Rice et al.[35]

4. Conclusion

The electronic emission spectrum of the $A^1\Pi$–$X^1\Sigma^+$ system of AlH has been recorded at high resolution with a Fourier-transform spectrometer. The present measurements of the line positions of the 0–0 and the 1–1 bands are more precise than those previously available.[36,40] The present data were combined with infrared vibration–rotation measurements of the fundamental 1–0 band of AlH[32] and the $J = 1$–0 pure rotational transition[42] to extract improved molecular constants for the $A^1\Pi$ state of AlH.

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Table 3. Equilibrium Constants (cm⁻¹) for the $A^1\Pi$ State of AlH

Constants^a	$A^1\Pi$
B_e	6.38632(14)
α_e	0.73198(27)
$10^4 \times D_e$	3.772(55)
$10^4 \times \beta_e$	5.12(11)
$\Delta G(1/2)$	1082.7768(16)
r_e (Å)	1.648328(18)

^{a}Numbers in parentheses are one standard deviation in the last digit.

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