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THEORETICAL CALCULATIONS OF H(2) CARS SPECTRA FOR PROPELLANT FLAMES

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DECEMBER 1983



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REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER Technical Report ARLCD-TR-83048	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) THEORETICAL CALCULATIONS OF H(2) CARS SPECTRA FOR PROPELLANT FLAMES		5. TYPE OF REPORT & PERIOD COVERED
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) Joanne Fendell L. E. Harris Kenneth Aron		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS ARDC, LCWSL Applied Sciences Div [DRSMC-LCA-G(D)] Dover, NJ 07801		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 1L16122AH60
11. CONTROLLING OFFICE NAME AND ADDRESS ARDC, TSD STINFO Div [DRSMC-TSS(D)] Dover, NJ 07801		12. REPORT DATE December 1983
		13. NUMBER OF PAGES 29
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE
16. DISTRIBUTION STATEMENT (of this Report) Approved for public release; distribution unlimited.		
17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)		
18. SUPPLEMENTARY NOTES		
19. KEY WORDS (Continue on reverse side if necessary and identify by block number) CARS Vibrational raman Propellant Hydrogen Rotational raman		
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) High energy pure rotational H(2) CARS transitions have recently been observed in several spectral regions, most notably, the NO and CO CARS regions. H(2) CARS is of interest because hydrogen is a major combustion product of propellants, especially nitramine propellants. For this reason, calculations were performed to assign the observed H(2) transitions. <p style="text-align: right;">(cont)</p>		

20. ABSTRACT (cont)

For this particular case, the calculations of interest are for the observed S- and Q-branch transitions of H(2) for v equal to 0 and 1. Spectroscopic constants available in the literature and the results of an ab initio calculation performed by Ermler were used in the analysis.

Ermler used the potential energy curves for the ground state of hydrogen calculated by Kolos and Wolniewicz over a range of $0.4 \leq R \leq 10.0$ a.u. as the basis for this calculation. The result is a set of spectroscopic constants superior to any other set examined.

Having assigned the transitions, the third order susceptibility, which gives a model for data reduction of CARS spectra can be calculated. Temperature and concentration for a given species can be obtained by comparing the theoretical spectra with experimental results.

ACKNOWLEDGMENT

The authors would like to thank Professor W. C. Ermler, Stevens Institute of Technology, for providing us with the ab initio calculations and helpful discussions.

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INTRODUCTION

The use of coherent anti-stokes Raman scattering (CARS) spectroscopy in combustion studies is well documented (refs 1 through 5). Hydrogen is a major combustion product of propellants, as illustrated by H₂ CARS S-branch transitions, obtained from both CH₄/N₂O model propellant flames and actual propellant flames (refs 6 and 7).

Because the use of H₂ CARS spectra to obtain temperature and concentration profiles is limited by the reliability of the spectroscopic constants, a literature search was conducted (refs 8 through 11) to find a suitable set of spectroscopic constants, and a set of ab initio calculations were obtained from Ermler.¹ The method used by Ermler is described briefly below. A more complete treatment is given in the literature (refs 12 through 15). Overall the transition energies calculated on the basis of Ermler's constants agreed most closely with our experimental results, so that particular set of spectroscopic constants was used to calculate H₂ S- and Q-branches.

CARS THEORY

The observed CARS spectrum is proportional to the square of the modulus of the third order susceptibility, $\chi^{(3)}$, which is the sum of a resonant term χ_r , and a nonresonant term χ_{nr} , which are related to the vibrational and electronic displacement, respectively

$$\chi^{(3)} = \chi_r + \chi_{nr} \quad (1)$$

The resonant term is calculated as the sum of Lorentian line shapes of each rotational transition

$$\chi_r = \sum_j K_j \frac{\Gamma_j}{(2\Delta\omega_j - i\Gamma_j)} \quad (2)$$

given that

$$K_j = \frac{2N}{h} |\alpha_j|^2 (\Delta p_j^{(0)}) \Gamma_j^{-1} \quad (3)$$

where N is the number density, α_j is the isotropic polarizability matrix element for the transition, $\Delta p_j^{(0)}$ is the normalized population difference between the

¹ Personal communication between W. C. Ermler, Stevens Institute of Technology, Hoboken, NJ and J. Fendell, ARDC, May 1983.

molecular energy levels involved in the transition, ω_j is the isolated pressure-broadened linewidth, and $\Delta\omega_j = \omega_1 - \omega_2 - \omega_j$. The calculated $|\chi^{(3)}|^2$ is first convoluted over the laser shapes, then over a triangular slit function.

χ_r is the sum of real and imaginary components χ' and χ'' , respectively, so that

$$|\chi^{(3)}|^2 = \chi'^2 + 2\chi'\chi_{nr} + \chi''^2 + \chi_{nr}^2 \quad (4)$$

χ' and χ'' display dispersive and resonant behavior with respect to the detuning frequency, $\Delta\omega_j$.

As the concentration of the resonant species is lowered, the cross term $2\chi'\chi_{nr}$, which is dispersive, influences the shape of the spectrum. The observation of dispersively modulated spectra allows temperature and concentration to be estimated on the basis of model calculations.

The concentration of various species from the ratio of the total CARS intensity to the nonresonant intensity at any frequency where resonant transition of the species occurs can also be estimated. In broadband CARS, the nonresonant susceptibility is usually observed directly from regions where no resonance occurs. The spectral distribution of the nonresonant susceptibility, which reflects that of ω_2 , can be obtained either from measurements of the distribution of ω_2 or directly from measurements of a nonresonant gas (ref 16).

RESULTS

Spectroscopic Constants

In an effort to find theoretical support for experimental observations, a literature search for spectroscopic constants for the $X^1\Sigma^+$ state of hydrogen was conducted. Constants published by Stoicheff (ref 8), Fink et al. (ref 9), Herzberg (ref 10), and Huber and Herzberg (ref 11) were available in the literature. Another set was obtained by data reduction of the results obtained from Ermler.

In his calculations, Ermler used potential energy curves for the $X^1\Sigma^+$ state of hydrogen calculated by Kolos and Wolniewicz (ref 13). These potential^g curves are obtained by using a generalized James-Coolidge wavefunction with variational parameters in elliptic coordinates, which is then used to solve the Schrodinger equation by numerical methods. Double precision arithmetic using 15 vibrational and 15 rotational levels were used in the calculations.

Application of Spectroscopic Constants

The total energy of a molecule is a function of both the rotational and vibrational quantum numbers. The expression is given as

$$E(v,J) = \omega_e(v + 0.5) - X_e \omega_e(v + 0.5)^2 + Y_e \omega_e(v + 0.5)^3 - Z_e \omega_e(v + 0.5)^4 + B_v(J^2 + J) - D_v J^2(J + 1)^2 + H_v(J^3)(J + 1)^3$$

where

$$B_v = B_e + \alpha_e(v + 0.5) + \gamma_e(v + 0.5)^2$$

$$D_v = D_e + \beta(v + 0.5) + \delta_e(v + 0.5)^2$$

$$H_v = H_0 + H_1(v + 0.5) + H_2(v + 0.5)^2$$

It is useful to know the values of B_v , D_v , and H_v , since they allow us to predict the frequencies at which pure rotational (S-branch) and ro-vibrational (Q-branch) transitions will occur.

$$\text{S-branch} = E(I,J+2) - E(I,J)$$

$$\text{Q-branch} = E(I+1,J) - E(I,J)$$

In general, B_v , D_v , and H_v are approximated by second-order polynomials. Stoicheff, Herzberg, and Huber and Herzberg stated these polynomials explicitly, but Fink et al. and Ermler stated these quantities at various values of v . In order to use these values in a systematic way in calculating S-branch and Q-branch spectra, values of B_v , D_v , and H_v corresponding to $v = 0,1,2$ were fitted to a second-order polynomial. The estimated standard error for these correlations ranged between 10^{-3} and 10^{-18} for Fink's results and between 10^{-11} and 10^{-18} for Ermler's results. The values for B_v , D_v , and H_v from all sources are compiled in table 1.

In table 2, the calculated S-branch frequencies are compared to our recently obtained experimental results. There is a clear difference among the results obtained by using various constants to calculate higher S-branch transitions. The constants of Fink et al. and Ermler were used to calculate the transition frequencies found in tables 3 and 4, since these particular sets of constants agreed most closely with experimental results.

Calculation of H_2 CARS S- and Q-Branches

H_2 CARS S- and Q-branches were calculated using Ermler's spectroscopic constants. The Q-branch results for various temperatures are given in figures 1, 2, and 3. The S-branch results shown in figure 4 for the $S_0(7,5)^2$ transition at 1815 cm^{-1} illustrates conditions comparable to those in the $\phi = 1.8 \text{ CH}_4/\text{N}_2\text{O}$ flame

² S-branch transitions are labeled using the notation $S_v(J', J'')$.

in which it was observed (refs 6 and 7). In these calculations, a Doppler broadened linewidth and a 3.0 cm^{-1} slit function were assumed.

DISCUSSION

The recently observed higher S-branch transitions (refs 6 and 7) are compared to the transition frequencies given by various spectroscopic constants in table 2. In all cases, the constants of Fink et al. and Ermler agree more closely with experimental results than other constants. For the $v = 0$ transitions, the results of Fink et al. and Ermler diverge as the transitions increase in frequency. Ermler's results are closer to the experimental results for $S_0(9,7)$ than those of Fink et al. For $S_0(11,9)$, the difference between experimental results and those of Fink et al. is large enough to be experimentally discernible. The frequency of the $S_0(11,9)$ transition has been observed experimentally at 2131 cm^{-1} by us (refs 6 and 7) as well as Farrow et al. (ref 17) in good agreement with Ermler's value of 2130 cm^{-1} . The results obtained for $v = 1$ do not differ greatly for either set of constants until levels higher than the observed transitions are reached. On the basis of present experimental data, Ermler's constants agree most closely with the experiment.

For both $v = 0$ and $v = 1$, Ermler's constants predict that the Q-branch transitions will be more closely spaced than the results of Fink et al. As seen from the computed results (table 3), Ermler's constants predict experimentally discernible differences from those at Fink et al. (ref 18) beginning at $J''=6$. The computed CARS Q-branch spectra (fig. 1 and 1a) show that above 2500 K, Q-branches above $J''=6$ are significantly populated. Q-branch spectra will be taken to confirm the validity of the spectroscopic constants at these temperatures.

The results obtained indicate that the set of spectroscopic constants obtained from the ab initio calculations are in better agreement with experimental results than other sets of constants.

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Table 1. Comparison of spectroscopic constants (cm^{-1})

<u>Constants</u>	<u>Herzberg</u>	<u>Huber and Herzberg</u>	<u>Stoicheff</u>	<u>Fink et al.</u>	<u>Ermler</u>
Be	60.800	60.853	60.840	60.8318	60.7922
α_e	-2.993	-3.062	-3.0177	-3.0087	-3.0320
β_e	0.025	0.057	0.0285	0.0266	0.0350
De	0.0464	0.00471	0.04684	0.0471	0.0448
Be	-0.00134	-0.0027	-0.0017	-0.0029	-0.0016
δ_e	--	0.0004	3×10^{-5}	4.5×10^{-4}	4.5×10^{-5}
H_0	5.18×10^{-5}	--	5.2×10^{-5}	5.62×10^{-6}	3.23×10^{-5}
H_1	--	--	--	-1.7×10^{-6}	-8.70×10^{-7}
H_2	--	--	--	4×10^{-7}	-2.50×10^{-8}
ω_e	4395.2	4401.21	4401.21	4401.217	4400.39
$\chi_e \omega_e$	117.90	121.33	121.43	121.343	120.814
$y_e \omega_e$	0.29	0.812	0.892	0.8145	0.7241
$Z_e \omega_e$	0.	0.	0.	0.	0.

Table 2. Calculation of selected transitions (cm^{-1})

<u>Transition^a</u>	<u>Experimental^b</u>	<u>Stoicheff</u>	<u>Fink et al.</u>	<u>Ermler</u>	<u>Huber and Herzberg</u>
$S_0(7,5)$	1446	1447.67	1447.61	1447.80	1449.83
$S_0(9,7)$	1809	1817.95	1817.18	1815.08	1822.05
$S_0(11,9)$	2131	2145.01	2141.91	2130.32	2150.30
$S_1(9,7)$	1721	1725.42	1719.90	1721.81	1723.13
$S_1(11,9)$	2020	2035.96	2019.02	2019.83	2032.29

^a The transitions are labeled " $S_V(J',J'')$."

^b The experimental data were taken from CARS data given in refs 7 and 18.

Table 3. Computed energies of H₂ Q-branch transitions

J	V = 0		V = 1	
	<u>Fink et al.</u>	<u>Ermler</u>	<u>Fink et al.</u>	<u>Ermler</u>
0	4161.18	4161.12	3925.82	3927.00
1	4155.25	4155.20	3919.99	3920.23
2	4143.32	4143.40	3908.16	3908.71
3	4125.21	4125.79	3890.00	3891.52
4	4100.68	4102.49	3865.00	3868.80
5	4069.40	4073.65	3832.52	3840.61
6	4030.94	4039.43	3791.70	3807.21
7	3984.82	4000.08	3741.56	3768.80
8	3930.44	3955.85	3680.93	3725.63
9	3867.15	3907.04	3608.47	3677.99
10	3794.19	3853.97	3522.70	3626.20
11	3710.74	3797.03	3421.95	3570.64
12	3615.89	3736.61	3304.38	3511.69
13	3508.63	3673.17	3168.01	3449.80
14	3387.88	3607.17	3010.67	3385.44
15	3252.49	3539.16	2830.03	3319.12
16	3101.21	3469.66	2623.60	3251.37
17	2932.70	3399.29	2388.70	3182.80
18	2745.56	3328.67	2122.53	3113.99
19	2538.28	3258.47	1822.07	3045.63
20	2309.30	3189.39	1484.18	2978.39

Table 4. Computed energies of H₂ S-branch transitions

<u>J'</u>	<u>J''</u>	<u>V = 0</u>		<u>V = 1</u>	
		<u>Fink et al.</u>	<u>Ermler</u>	<u>Fink et al.</u>	<u>Ermler</u>
0	0	354.37	354.13	336.69	336.41
3	1	587.02	586.74	557.68	557.33
4	2	814.40	814.22	773.55	773.30
5	3	1034.65	1034.67	982.47	982.50
6	4	1246.16	1246.37	1182.79	1183.26
7	5	1447.61	1447.80	1373.12	1374.15
8	6	1638.11	1637.69	1552.39	1553.96
9	7	1817.18	1815.08	1719.90	1721.81
10	8	1984.89	1979.37	1875.37	1877.15
11	9	2141.91	2130.32	2019.02	2019.83
12	10	2289.56	2268.18	2151.61	2150.14
13	11	2429.91	2393.64	2274.49	2268.85
14	12	2565.84	2507.95	2389.67	2377.27
15	13	2701.09	2612.97	2499.89	2477.29
16	14	2840.35	2711.12	2608.65	2571.43
17	15	2989.33	2805.54	2720.27	2662.86
18	16	3154.84	2900.08	2839.97	2755.49
19	17	3344.84	2999.35	2973.92	2853.99
20	18	3568.49	3108.80	3129.26	2963.83
21	19	3836.30	3234.71	3314.22	3091.35

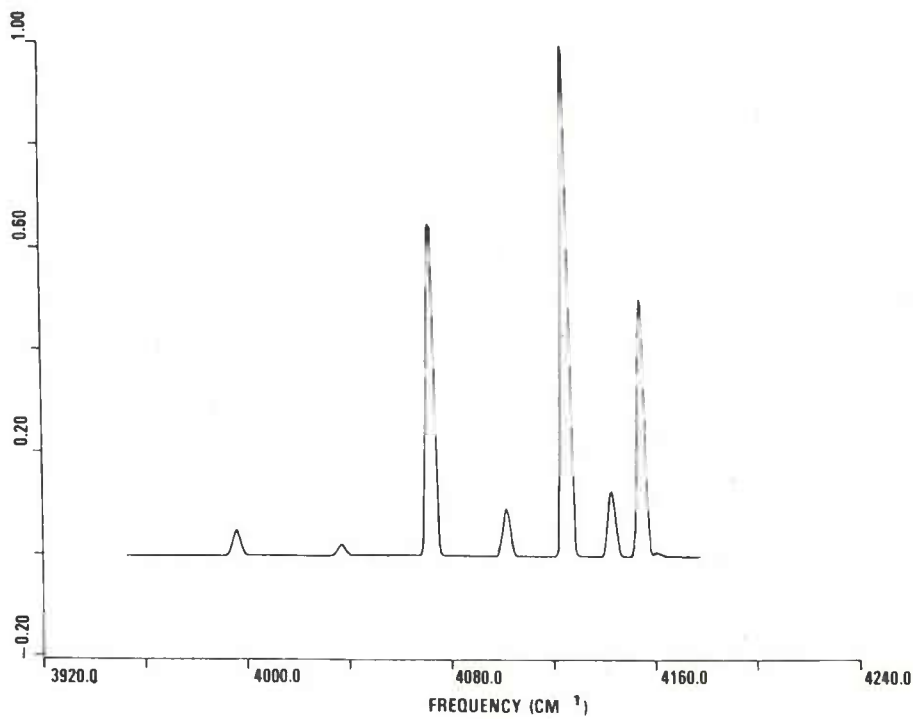


Figure 1. H₂ CARS Q-branch spectra at 2500 K

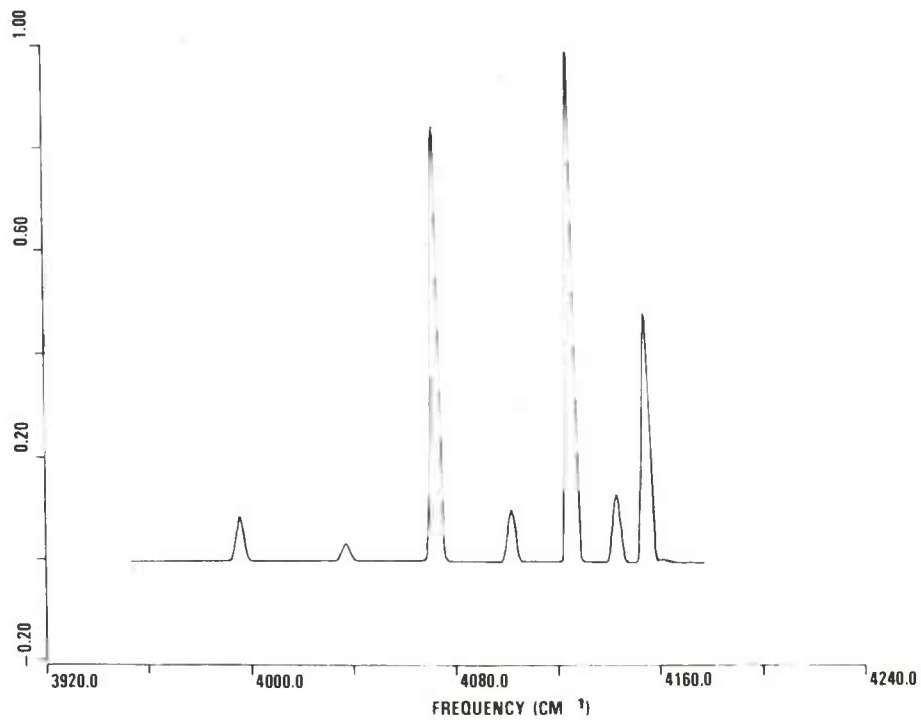


Figure 2. H₂ CARS Q-branch spectra at 3000 K

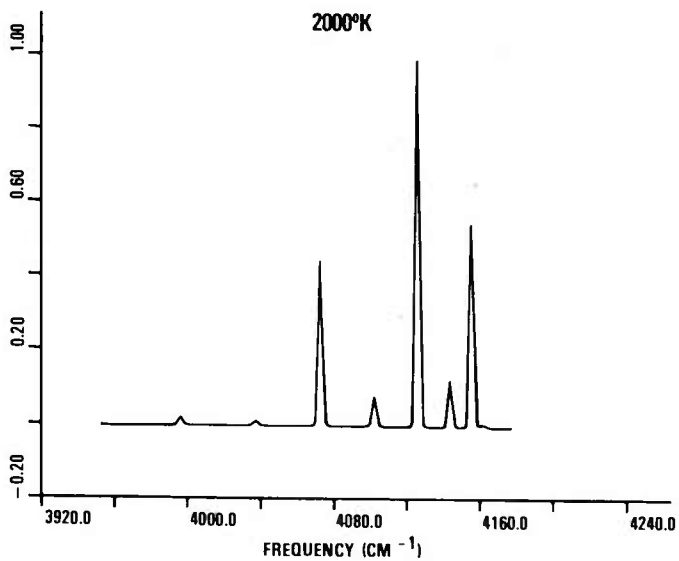
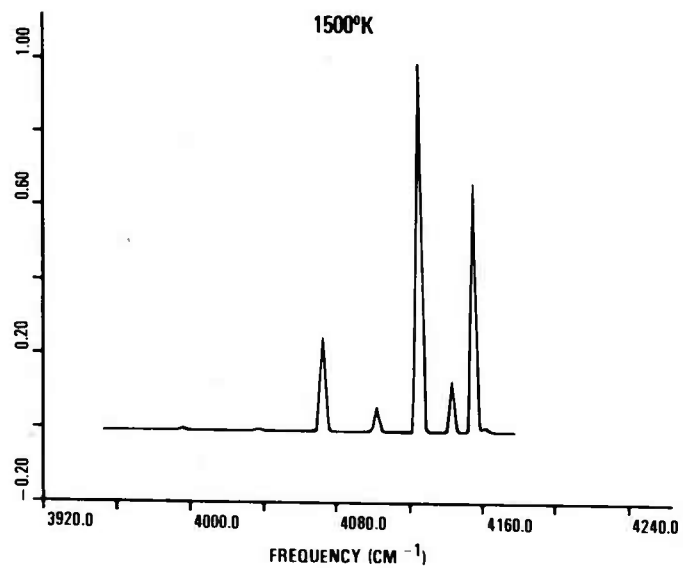
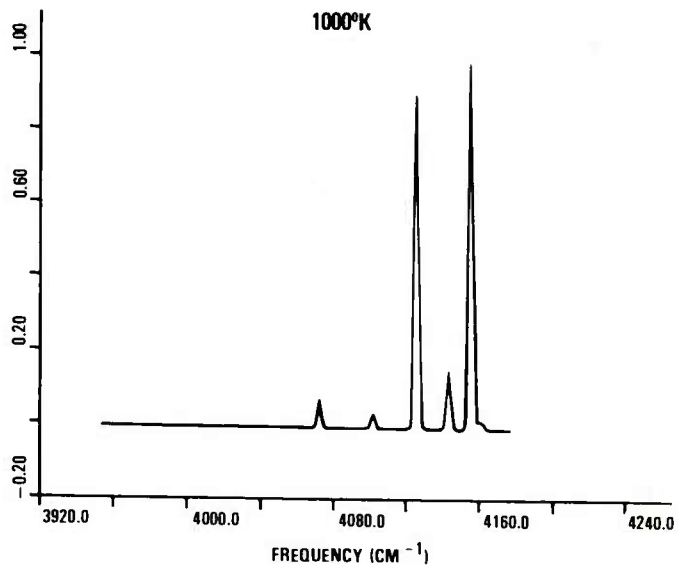
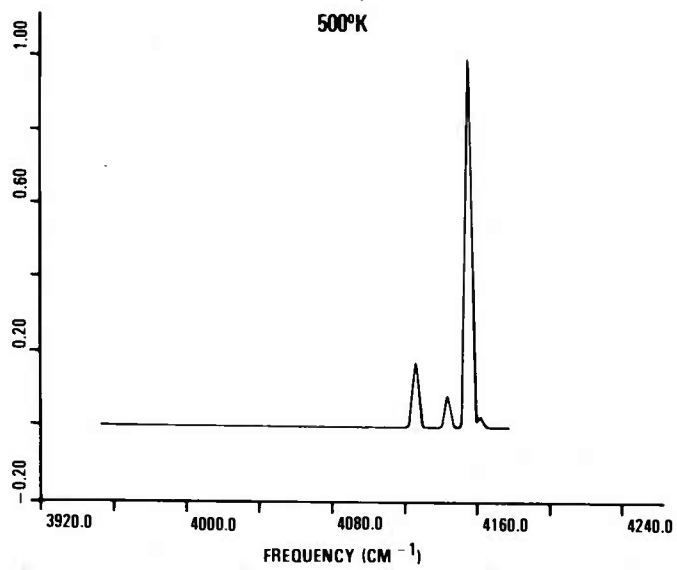


Figure 3. Temperature variation of H₂ CARS spectra

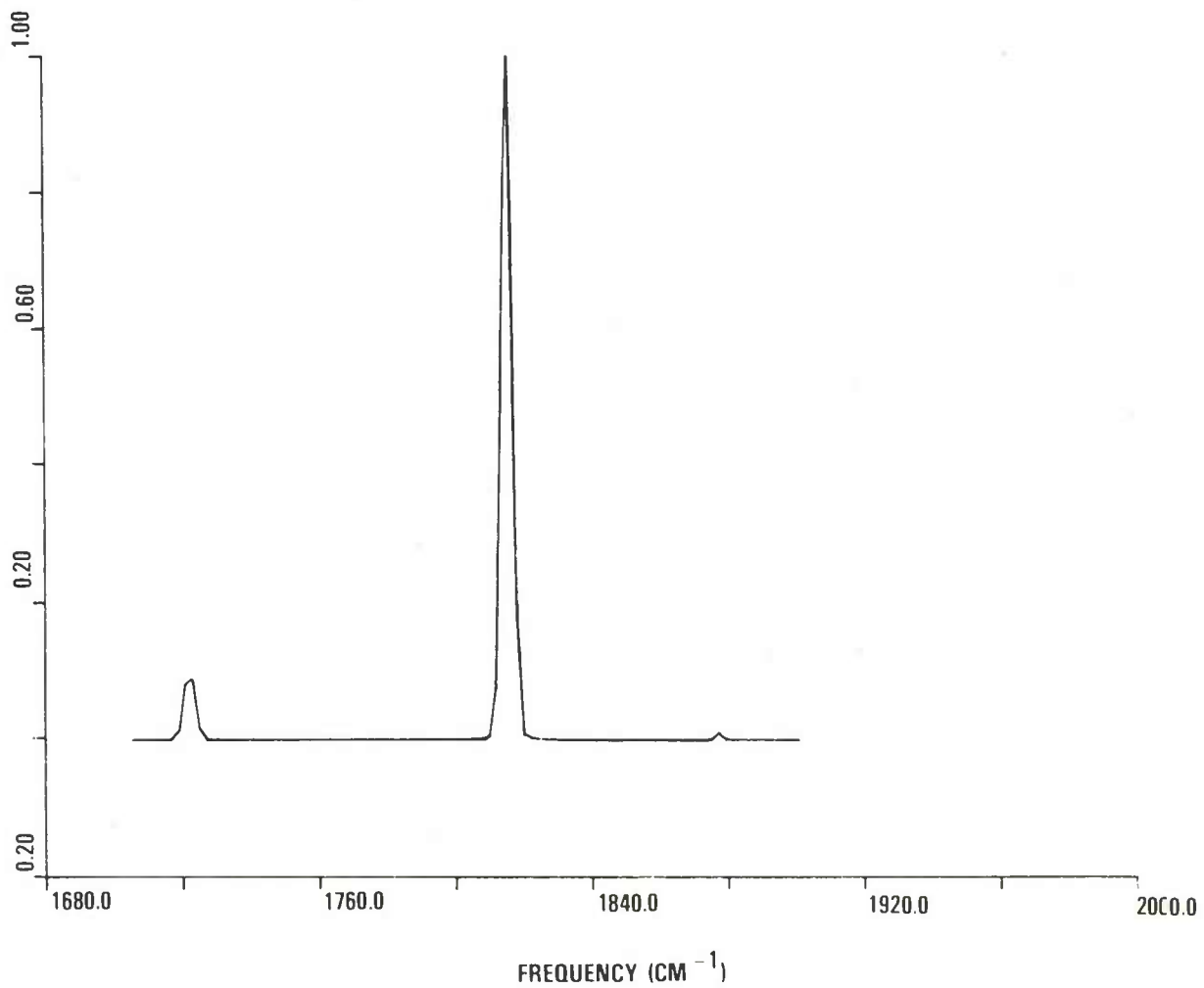


Figure 4. H₂ CARS S-branch at 2732 K

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