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DIODE LASER MEASUREMENTS ON DIDEUTEROACETYLENE

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ABSTRACT

Diode laser measurements on the $\nu_4 + \nu_5$ band of dideuteroacetylene are reported. Various lines of interest in this band have been measured to an accuracy of 0.001 cm^{-1} and the self pressure broadening and helium pressure broadening coefficients determined. These measurements are of relevance to the recent observation of optically pumped laser action in this molecule.

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1. INTRODUCTION

Strong optically pumped laser action has recently been obtained in dideuteroacetylene, $^{12}C_2D_2$, by pumping the $(\nu_4+\nu_5)$ combination band with a $^{12}C^{16}O_2$ TEA laser (H.N. Rutt and J.M. Green 1978). We report here accurate spectroscopic measurements on some C_2D_2 lines together with pressure broadening and absolute absorption coefficients. These parameters are of importance to the operation of the C_2D_2 laser.

2. EXPERIMENTAL

A commercial current tuned diode laser was used to obtain absorption spectra of C_2D_2 in the 9.5 μm region, with a resolution limited only by the Doppler width, 74 MHz. A germanium etalon provided fringes spaced at 0.049 cm^{-1} and absolute calibration was obtained by including in the optical path a removeable 1 metre long cell containing 7 Torr of CO_2 at ~ 200°C. The 1m long C_2D_2 cell could be cooled to 77 K.

The C_2D_2 used in these experiments was obtained commercially, with a stated isotopic purity of 99.5%. The only impurities detected in low resolution infrared spectra were traces of C_2HD, D_2O and very small amounts of C_2H_2 and HDO.

3. SPECTRAL DATA

The absorption spectrum of C_2D_2 in the 9.5 μm region has previously been investigated in great detail (S.C. Hurlock, S. Ghersetti and K.N. Rao 1971; S. Ghersetti, J. Pliva and K.N. Rao 1971; A. Baldacci, S. Ghersetti and S.C. Hurlock 1972) at a resolution of 0.04 cm^{-1}. The spectrum previously reported consists of the combination band $(\nu_4+\nu_5)\Sigma_u^+$ arising from the Σ_g^+ ground state overlapped by the hot bands $2\nu_4+\nu_5-\nu_4$ and $2\nu_5+\nu_4-\nu_5$. Numerous multiple hot bands occur weakly. Absorption to the $(\nu_4+\nu_5)\Delta_u$

level is formally forbidden ($\Delta l = 2$), but data on this level are available from hot band measurements reported in (S. Ghersetti, J. Pliva and K.N. Rao 1971).

Table 1 shows a number of C_2D_2 lines measured with an accuracy of 0.002 - 0.015 cm^{-1}, the accuracy being limited by the accuracy to which the etalon fringe spacing is known. The CO_2 frequencies are taken from a line list for the CO_2 laser band given in (J. Dupre-Maquaire and P. Pinson 1976), based on the constants given in (F.R. Peterson, D.G. McDonald, J.D. Cupp and B.L. Danielson 1973).

The first three transitions listed are of particular interest since laser action has been obtained in C_2D_2 pumped by the CO_2 lines used here as frequency calibrations (H.N. Rutt and J.M. Green 1978). Figs 1 and 2 show the spectra recorded for two of these lines. The three laser pump lines have offsets of 0.031, 0.032 and 0.011 cm^{-1}. The third and fourth transitions in Table 1 are assigned to the transition from the ground state to the level $(\nu_4+ \nu_5)\Delta_u$, which has not previously been observed. These weak absorption features persisted on cooling the gas to 150 K, and then decreased in intensity (approximately in proportion to nearly Σ_u^+ transitions of similar J) as the gas condensed. This conclusively demonstrates that they are not hot band lines, and furthermore they do not coincide with the lines of $^{13}C^{12}CD_2$ given in (K.P. Pollard, S. Ghersetti and K.N. Rao 1969). Whilst the ground state to $(\nu_4+ \nu_5)\Delta_u$ transition is forbidden, an ℓ-type resonance exists between the Σ_u^+ and Δ_u components of $\nu_4+ \nu_5$ (A. Baldacci, S. Ghersetti and S.C. Hurlock 1972). This resonance is responsible for the appreciable transition moment from the ground state to Δ_u.

Although the primary interest in diode measurements of the C_2D_2 spectrum was the lines known to give rise to laser action, we include in Table 1 a number of other lines which have been measured with high accuracy. In most cases these agree with earlier measurements to within 0.02 cm^{-1}, confirming the excellent accuracy of the spectroscopic constants for C_2D_2 given in (S.C. Hurlock, S. Ghersetti and K.N. Rao 1971; S. Ghersetti, J. Pliva and K.N. Rao 1971; A. Baldacci, S. Ghersetti and S.C. Hurlock 1972).

Several other pump lines have been observed in C_2D_2 but unfortunately the tuning characteristics of the diodes available did not permit measurements to be made on these lines.

4. PRESSURE BROADENING

Pressure broadening coefficients are of considerable importance in the operation of optically pumped gas lasers. Measurement of this parameter permits estimates of the permissible pump detuning to be made, and also allows calculation of the extent of rotational relaxation occurring under laser conditions. Pressure broadening coefficients have not previously been reported for C_2D_2.

Fig.3 shows the dependence of the linewidth of the R(6)$(\nu_4+\nu_5)\Sigma_u^+$ line on pressure for self broadening and helium broadening at room temperature. The R(6) transition was chosen as it is well clear of nearby hot band lines, and was in a good tuning region for the diode laser. In order to measure the self broadening coefficient a very short path length cell (8.1 mm) had to be used to obtain a suitable line centre absorption.

The self broadening and helium broadening coefficients obtained from Fig.3 are 12.4 and 3.6 MHz/Torr full width at half maximum respectively. The absolute absorption coefficient for the R(6) line at 300 K was 0.84 $\pm$ 0.1 cm^{-1} under fully pressure broadened conditions ($\geq$ 20 Torr). This shows that under typical laser conditions (1 metre path, detunings $\sim$ 0.03 cm^{-1} and pressures of up to 65 Torr) total absorption of the pump beam will occur.

5. CONCLUSIONS

The use of a current tuned diode laser has permitted the measurement of a number of parameters of importance to the C_2D_2 laser. Similar measurements on other optically pumped gases would be very useful, especially to obtain spectroscopic assignments for the pump transitions in heavier molecules such as CF_4.

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TABLE 1 DIODE MEASUREMENTS ON C_2D_2

C_2D_2 Line			Measured cm^{-1}	Relative to CO_2		Calculated from Ref. (2,3,4)
$\nu_4+\nu_5\ \Sigma_u^+$	R(0)		1043.194 ± 0.002	P(24)	1043.163	1043.199
" Σ_u^+	P(6)		1031.445 ± 0.001	P(36)	1031.477	1031.447
" Δ_u	R(16)		1078.582 ± 0.002	R(20)	1078.591	1078.58
" Σ_u^+	P(11)		1023.283 ± 0.005	P(44)	1023.189	1023.298
" Δ_u	R(19)		1084.24 ± 0.015	R(28)	1083.479	1084.25
" Σ_u^+	R(3)		1048.35 ± 0.01	P(18)	1048.661	1048.380
$2\nu_4+\nu_5-\nu_4$						
$\pi_u-\pi_g$	R(21)	d/d	1081.138 ± 0.002	R(24)	1081.087	1081 139
$2\nu_5+\nu_4-\nu_5$						
$\pi_g-\pi_u$	R(3)	d/d	1043.104 ± 0.002	P(24)	1043.163	1043.123
" "	R(3)	c/c	1043.016 ± 0.006			1043.033

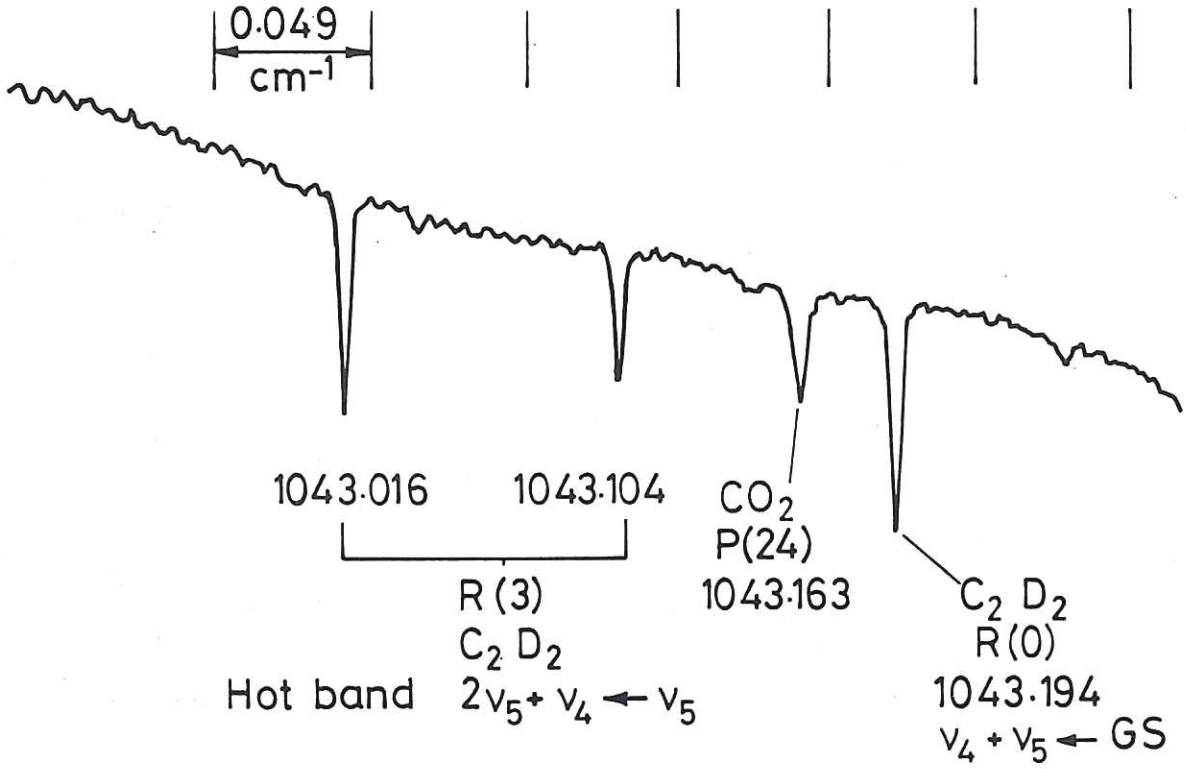

Fig.1 Diode laser spectrum of C_2D_2 in the region of R(0).

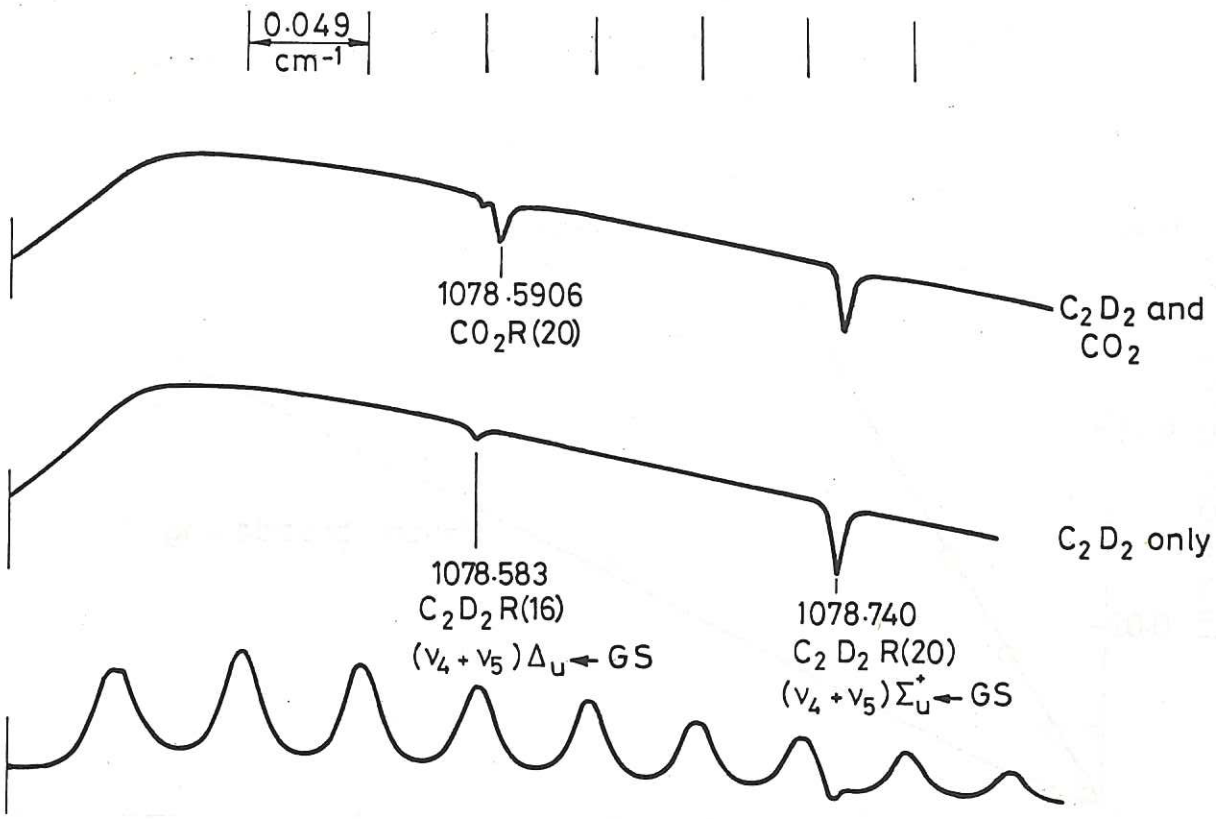

Fig.2 Diode laser spectrum of C_2D_2 in the region of R(20).

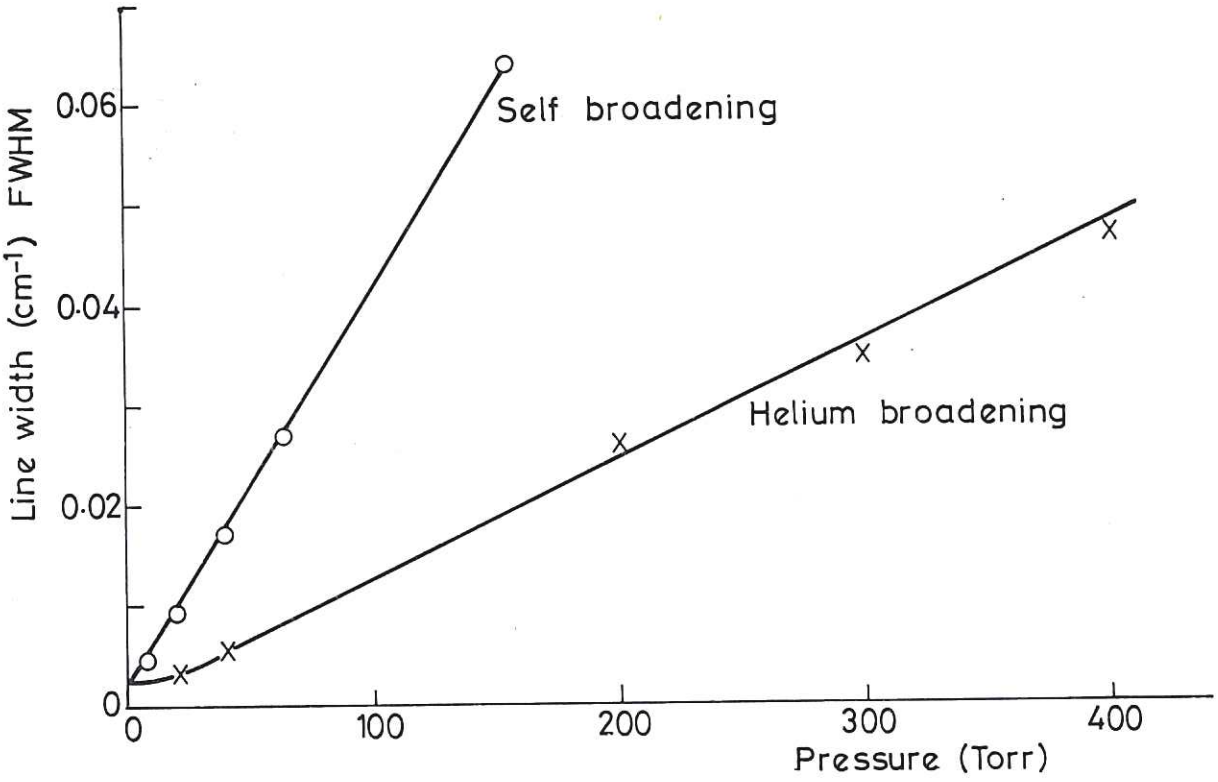

Fig.3 Pressure broadening of C_2D_2 for R(6) of $\upsilon_4 + \upsilon_5$ (Σ_u^+).