

High Resolution Lifetime Studies of the $d^3\Pi_g$, $C^1\Pi_g$ and $D^1\Sigma_u^+$ States in C_2 with Applications to Estimates of the Solar Carbon Abundance

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Abstract

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Lifetimes have been measured for a number of vibrational levels of the $d^3\Pi_g$, $C^1\Pi_g$ and $D^1\Sigma_u^+$ states in the C_2 molecule using the High Frequency Deflection technique. Particular attention was paid to various rotational levels of the $v'=6$ level of the $d^3\Pi_g$ state but no decrease in lifetime was found indicating the absence of earlier suggested predissociation effects, which would be an explanation of the occurrence of the high-pressure carbon bands. For the $d-a$ (Swan) bands oscillator strengths are found which are considerably higher than earlier estimates demanding a 40% decrease in the estimate of the solar carbon abundance.

1. Introduction

The spectrum of the C_2 molecule is one of the best studied among the fundamental molecules. Compilations of known data for this molecule may be found for instance in refs. [1, 2] while Fig. 1 shows a simplified diagram of the potential curves for some low-lying excited electronic states. The best known system of the C_2 molecule (the Swan bands) consists of transitions between the $d^3\Pi_g$ and the $a^3\Pi_u$ states where the latter state was long believed to be the ground state, but was later shown [1] to be situated 610 cm^{-1} above the ground state $X^1\Sigma_g^+$. The Swan bands are frequently observed in the spectrum of the sun as well as of carbon stars, comets and planet atmospheres. Indeed the present knowledge of the solar spectrum includes more than 40 rotational lines of the (0, 0) Swan band ($\lambda = 5165\text{ \AA}$) the majority of which are suitable for measurements of equivalent widths (cf. refs. [3, 4]). Hence the Swan band rotational components are very suitable for an estimate of the solar carbon abundance. Most atomic transitions used for the same purpose are either forbidden transitions [5–7] for which the abundance estimates have to rely upon theoretical oscillator strength (f -value) calculations [2], or a few transitions in the EUV region [33, 34].

Even a determination of the carbon abundance from the C_2 Swan bands is dependent upon the accuracy of the associated f -values for which we find experimental values between $f(0, 0) = 0.00014$ [10] up to $f(0, 0) = 0.025 \pm 0.004$ [11] in the literature. Lifetime investigations of the same (0, 0) band (see Table I) have yielded values between 778 ns and 170 ns corresponding to f -values between 0.0043 ± 0.0001 and 0.0178 ± 0.0018 . In view of these large discrepancies and the particular importance of this

oscillator strength it was decided to perform detailed lifetime investigations of several of the Swan bands as well as other low-lying states in the C_2 molecule using our new high resolution technique (the HDF technique [14–16]).

Another reason for the present investigation is the following. The determination of the carbon abundance from the C_2 lines is strongly dependent upon the dissociation energy D_0 for the molecule (cf. eq. (8)). For a long time $D_0(C_2)$ has been rather uncertain, one group of investigations have indicated a value 6–7 eV while other evidences have yielded a considerably lower value or 3.4–3.6 eV [17]. The main reason for assuming the lower value is the existence of the well known high pressure bands in C_2 (i.e. the Swan bands (6, v'')) the origin of which has been suggested [17] to be a consequence of predissociations of the $v'=6$ ($d^3\Pi_g$) level. If this interpretation is correct, $D_0(C_2)$ should be less than the excitation energy of the $v'=6$ level and larger than that of the $v'=5$ level, which leads to the range 3.4–3.6 eV. The primary experimental indication that the $v'=6$ level might be predissociated is that the (6, v'') bands and no other Swan bands are observed in discharge tubes containing carbon monoxide at relatively high pressures, which suggests that dissociated CO molecules form new C_2 molecules via inverse predissociation through the $v'=6$ ($d^3\Pi_g$) level. Since direct measurement of radiative lifetimes is by far the most sensitive probe of predissociation effects, we decided to extend the lifetime studies of the C_2 Swan bands to include levels up to $v'=6$ to see if any predissociations are present. The onset of predissociation is namely immediately noted as a decrease in the measured lifetime and in this way predissociation probabilities as small as 10^4 s^{-1} can be detected and this high sensitivity has recently led to discoveries of previously unknown predissociation cases [18–20].

2. Experiment and Results

For the present purpose, the HFD technique was applied with C_2H_2 at 1–5 mtorr as a target gas which was bombarded with 7 keV 20 mA electrons. The resulting emission spectrum was observed with a 2 m Czerny-Turner scanning monochromator equipped with an EMI 6256 SQ photomultiplier. Figure 2 shows small portions of this spectrum in the vicinity of (0, 0) band heads of the $d^3\Pi_g-a^3\Pi_u$ (Swan) and $C^1\Pi_g-A^1\Pi_u$ (Deslandres-d'Azambuja) systems. At the present resolution (about 0.08 \AA FWHM) most of the rotational lines of the bands are resolved and an analysis is possible in order to ensure that no foreign lines appear in the spectra.

The lifetime measurements have most frequently been performed on the band heads, i.e. on a group of unresolved rota-

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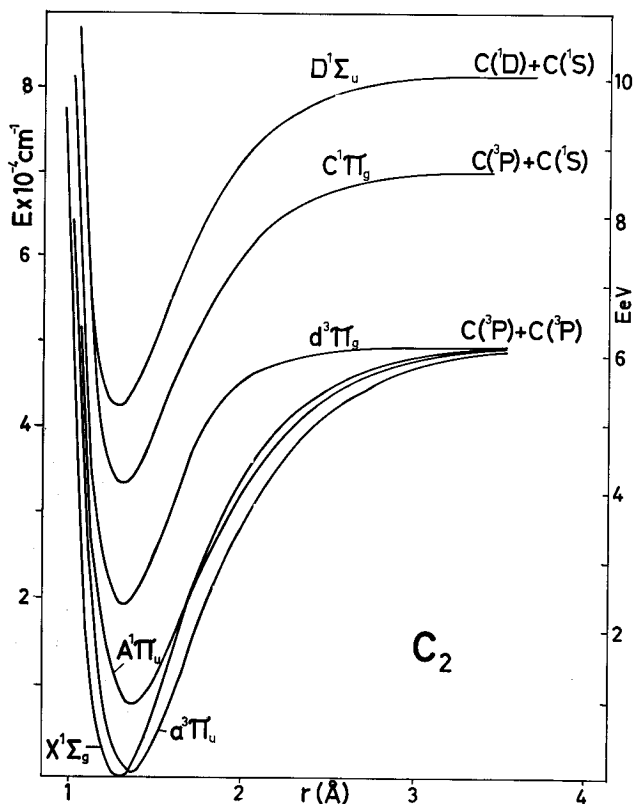


Fig. 1. Potential curves of some low-lying electronic states in the C₂ molecule.

tional lines originating from $J=7-22$ ($d^3\Pi_g$) and $J=3-15$ ($C^1\Pi_g$). For these two states the lifetime of a level v' has pervadingly been measured from the $\Delta v = +1$ bands (with the exception $\Delta v = 0$ for $v' = 0$) which appear with the highest intensities. No attempts have been made to resolve the dense structure of the $D^1\Sigma_u^+ - X^1\Pi_g$ (Mulliken) bands, only average lifetimes of the $\Delta v = 0$ sequence (i.e. $v' = 0-3$) have been extracted.

Table I. Lifetimes (ns) of various levels of the C₂ $d^3\Pi_g$, $C^1\Pi_g$ and $D^1\Sigma_u^+$ states

v'	$d^3\Pi_g$		$C^1\Pi_g$		$D^1\Sigma_u^+$
	Approximate J-interval	τ	Approximate J-interval	τ	
0	7-22	123 ± 6	3-15	31.1 ± 1.5	18.1 ± 1.0 ^a
1	7-22	124 ± 6	3-15	33.1 ± 1.5	
2	7-22	130 ± 6	3-15	32.6 ± 1.5	
3	7-22	128 ± 6	3-15	32.3 ± 1.5	
4	7-22	131 ± 6	3-15	36.1 ± 2.0	
5	7-22	137 ± 10			
6	7-22	140 ± 10			
6	> 50	147 ± 15			

Other experimental results:

$d^3\Pi_g$: $\tau(0) = 778$, $\tau(1) = 693$, $\tau(2) = 641$, $\tau(3) = 629$ [12]; $\bar{\tau} = 200 \pm 50$ [21]; $\bar{\tau} = 170 \pm 20$ [13]; $\bar{\tau} = 203 \pm 10$ [16],

$D^1\Sigma_u^+$: $\bar{\tau}(v=0-3) = 14.6 \pm 1.5$ [13].

^a Average value for $v' = 0-3$.

Examples of measured decay curves are shown in Figure 3. The displayed decay curves have been registered at 403.0 kHz electron beam sweep frequency (the d state) and 1 997 kHz (the C and D states) which are roughly the optimal sweep frequencies for these curves, i.e. the decays have been followed for 8-12 lifetimes. Recordings of the same curves have also been performed at 4 times lower frequencies (i.e. about 40 lifetimes) in order to search for possible cascade lifetimes. This test showed the absence of multiple exponentials i.e. that no cascades or blends are present in any of the cases. Actually it is obvious from the potential curves that essentially no cascades are expected in view of the selection rules. Thus, for instance, our results confirm that there are no $^3\Lambda_u$ states above the $d^3\Pi_g$ state which can feed the latter state. Tests of the pressure dependence showed that none of the studied lifetimes are affected by density-dependent effects in the range 1-5 mtorr.

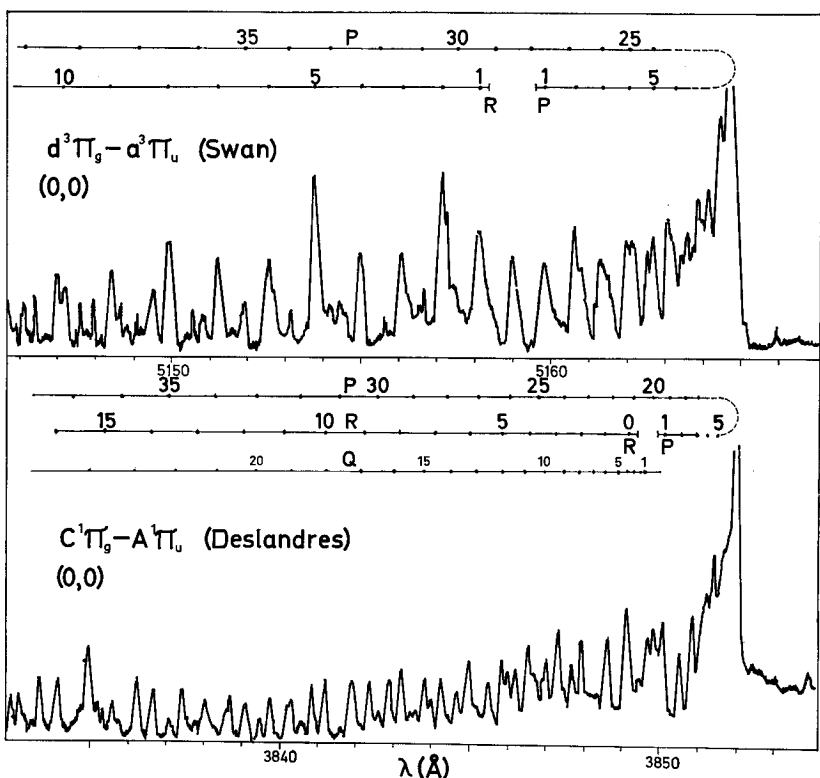


Fig. 2. Spectra of the (0,0) bands of the $d^3\Pi_g - a^3\Pi_u$ (Swan) and $C^1\Pi_g - A^1\Pi_u$ (Deslandres-d'Azambuja) systems in C₂ in the vicinity of the band-heads. The recordings have been performed using the High Frequency Deflection technique where C₂H₂ gas was bombarded with 7 keV, 20 mA electrons and the emission spectrum was studied using a 2 m Czerny-Turner scanning monochromator set at 0.08 Å FWHM resolution.

The results of our lifetime measurements are tabulated in Table I. For the $d^3\Pi_g$ state we find values between $\tau(v'=0) = 123 \pm 6$ ns and $\tau(v'=6, J > 50) = 147 \pm 15$ ns i.e., a barely discernible increase. Thus the $v'=6$ level cannot possibly be influenced by predissociation effects. More specifically, within the present error limits we find that a drop in $\tau(v'=6)$ of say 20 ns relative to $\tau(v'=5)$ would be easily detectable. This leads to an upper limit $A^{nr}(v'=6) < 10^8 \text{ s}^{-1}$ for the transition probability of a non-radiative transition from the $v'=6$ level. It would indeed be hard to explain quantitatively the creation of C_2 molecules via inverse predissociation through the $v'=6$ level of the $d^3\Pi_g$ state in view of this low transition probability. Thus other mechanisms should be searched for which could explain the appearance of the high pressure carbon bands and this problem should still be considered as unsolved. It should also be noted that Swan bands originating from $v' > 6$ are extremely weak, if present at all, in our spectral recordings at low pressure. This intensity drop also indicates the presence of unknown perturbations.

The results of the present lifetime investigations of the $v'=0-6$ levels of the $d^3\Pi_g$ state could be compared to results of earlier direct measurements as compiled in Table I. We find the new values to be 5-6 times lower than the results [12] obtained from fluorescence decay measurements using laser excitation and some 40% lower than other more recent measurements [13, 16, 21] which are essentially in mutual agreement. As a comment on the very long lifetime [12] it should be mentioned that this has been obtained via a fundamentally different excitation, namely by laser excitation of a vapor evaporated from a graphite block by the laser beam itself. However, it has been shown that such an evaporation yields predominantly C_3 molecules and not C_2 molecules (cf. ref. [22]) and the origin of the photons observed [12] at 6 Å band-pass might be questioned. The later investigations [13, 21] have been performed using the phase-shift technique with acetylene or benzene as target gases, while the value given in ref. [16] was a preliminary result obtained using the present technique. Even these later measurements [13, 16, 21] have been performed at a moderate spectral resolution (5-20 Å FWHM) and indeed we are able to roughly reproduce the ~ 200 ns lifetime by lowering our present resolution to about 5 Å. Thus we feel that this resolution is insufficient in lifetime measurements of the rather dense C_2 bands (cf. Fig. 2) and that even a 5 Å band-pass will in general include foreign spectral lines. However, we feel sure that the present resolution, which enables a detailed rotational analysis, is sufficient to exclude these blend effects revealing a 40% shorter lifetime (larger f -values) of the C_2 Swan bands.

Concerning the $C^1\Pi_g$ state no experimental lifetimes are reported, to our knowledge. We find a slight variation in $\tau(C^1\Pi_g, v')$ with v' in the range $\tau(0) = 31$ ns up to $\tau(4) = 36$ ns. Spectroscopic studies of this state have shown emission break-offs in the rotational series which have been interpreted as predissociation effects. These break-offs are found for instance at $J=103$ for $v'=0$ and $J=83$ for $v'=4$ [23] and the extrapolation to zero rotational energy has yielded a value for the dissociation energy $D_0^0(C_2) = (6.11 \pm 0.04)$ eV. The same authors [24] find similar values or 6.07 and 6.04 eV from extrapolations of the vibrational levels of the $d^3\Pi_g$ and $e^3\Pi_g$ states, respectively. It would be interesting to establish the predissociation effects in the $C^1\Pi_g$ state more accurately by lifetime investigations but unfortunately we find even the present resolution to be insufficient for studies of individual high rotational levels in view of the complexity of the Deslandres- d^2 Azambuja bands (cf. Fig. 2).

It is of interest to establish lifetimes and oscillator strengths for systems in C_2 which involve the ground state $X^1\Sigma_g^+$ and hence

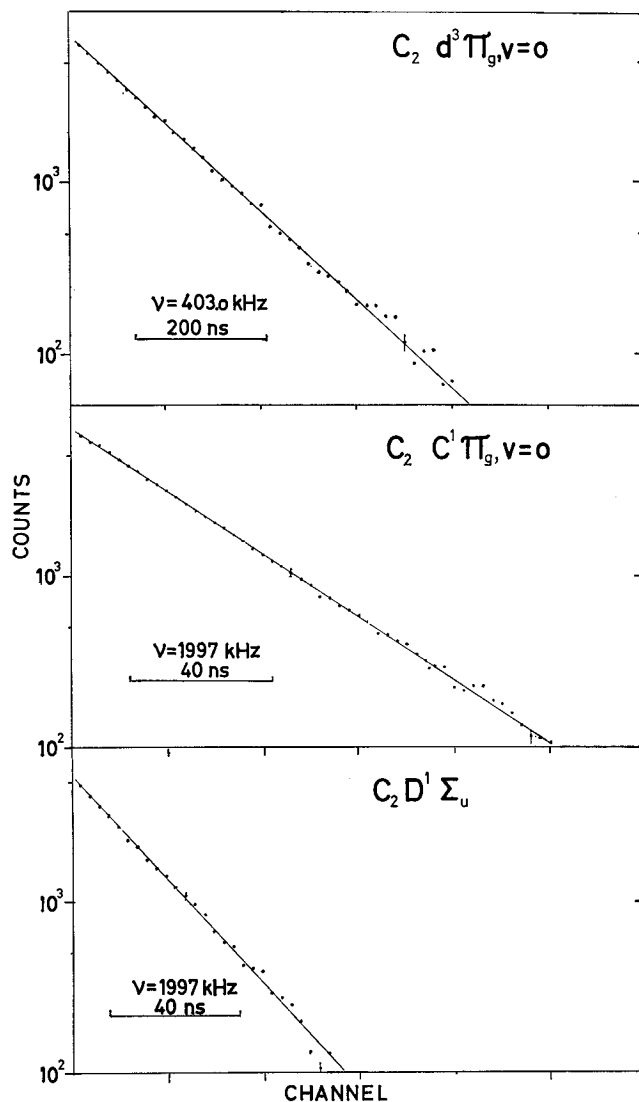


Fig. 3. Decay curves of rotational levels in the band-heads displayed in Fig. 2 as well as the $v=0-3$ levels of the $D^1\Sigma_u^+$ state. By applying different sweep frequencies, the individual lifetimes can be studied at optimum conditions yielding $\tau(d^3\Pi_g, v=0) = 123 \pm 6$ ns, $\tau(C^1\Pi_g, v=0) = 31.1 \pm 1.5$ ns and $\tau(D^1\Sigma_u^+, v=0-3) = 18.1 \pm 1.0$ ns.

could be used for abundance determinations of C_2 molecules in this state for instance in comets. Of the two known systems involving the ground state, the Phillips and Mulliken bands, the latter one appears in a more convenient spectral region (2 310-2 415 Å) and is also observed in our spectra. We have not made any attempts to resolve the dense structure of the Mulliken bands, and the lifetime has been measured on the unresolved group $\Delta v=0$ at 2 315 Å yielding the result $\tau(D^1\Sigma_u^+, v'=0-3) = 18.1 \pm 1.0$ ns.

3. The oscillator strengths of the Swan bands

The relation between the oscillator strength $f(v', v'')$ and the lifetime $\tau_{v'}$ of the upper level is (in c.g.s. units) given by

$$f(v', v'') = 1.499 \cdot \lambda^2(v', v'') \cdot \frac{g'}{g''} \cdot \frac{1}{\tau_{v'}} \cdot \frac{q(v', v'') \cdot \nu^3(v', v'') \cdot R_0^2(\bar{r}_{v', v''})}{\sum_{v''} q(v', v'') \cdot \nu^3(v', v'') \cdot R_0^2(\bar{r}_{v', v''})} \quad (1)$$

where $q(v', v'')$ is the Franck-Condon factor, $R_0(\bar{r}_{v', v''})$ is the electronic transition moment, $\bar{r}_{v', v''}$ the r -centroid and g' and g''

Table II. Oscillator strengths $f(v', v'') \times 10^3$ for various bands of the $d^3\Pi_g - a^3\Pi_u$ system in C_2 deduced from the present lifetime investigations

$v'' \dots$ v'	0	1	2	3	4	5	6
0	26.1	6.6	1.1	0.2	0.0	0.0	0.0
1	10.0	11.4	7.7	2.2	0.5	0.1	0.0
2	1.5	14.0	4.1	6.4	2.8	0.7	0.1
3	0.0	3.4	14.5	1.3	4.7	2.9	1.0

Other experimental results from absorption or shock-tube measurements: $f(0,0) \times 10^3 = 0.14$ [28]; 5 ± 3 [10]; 17 ± 4 [29]; 20.5 ± 9 [30]; 25 ± 4 [11].

are statistical factors ($g'/g'' = 1$ in the present case). In many cases R_0 can be considered as being constant in the summation in (1) and can in those cases be excluded from eq. (1). In the present case, however, the summation has to be carried out over several higher v'' levels before the summation in (1) converges because of the q factors. Hence we will make use of an experimentally determined variation [25] in R_0 for the Swan bands:

$$R_0^2(\bar{r}) = K(5.07 \cdot \bar{r}_A - 1)^2 \tag{2}$$

where K is a constant. Introducing (2), $\tau(0, 0) = 123 \pm 6$ ns and calculated Franck-Condon factors [26] and r -centroids [27] into (1) we obtain

$$C_2 \text{ Swan bands } f(0, 0) = (26.1 \pm 1.6) \times 10^{-3} \tag{3}$$

A calculation of $f(0, 0)$ from eq. (1) with $R_0^2 = \text{const.}$ yields $f(0, 0) = 0.0254$, i.e. this approximation gives a 3% error in the present case. Oscillator strengths for other Swan bands are calculated from (1) and (2) in the same way using the present lifetime results and they are given in Table II up to (3, 6).

Using the known relation between $\tau_{v''}$ and R_0^2 (in c.g.s. units, R_0 in atomic units):

$$1/\tau_{v''} = 2.026 \times 10^{-6} \cdot \sum_{v'''} q(v', v''') \cdot v'^3(v', v''') \cdot R_0^2(\bar{r}_{v', v'''}) \tag{4}$$

we get, using the same procedure as above for the (0, 0) band a value for the constant K in eq. (2) $K = 0.020$ a.u. In this connection it should be pointed out that some authors define eq. (2) as a lower state degenerate sum ($\Sigma R_0^2 = g' \cdot R_0^2$) in which case the constant becomes $g''K$.

The present value of $f(0, 0)$ (3) could be compared to other values obtained from absorption or shock-tube measurements as compiled in Table II. Obviously the extreme values differ a factor 180 while our new $f(0, 0)$ value is in agreement with the highest value [11] obtained in these other measurements. Adding the $f(0, 0)$ values which might be determined from the earlier lifetime measurements, an average value $f(0, 0) = 0.016 \pm 0.007$ is deduced from the previous investigations, i.e. a 40% lower value than given by (3).

4. The solar carbon abundance

Having established a 40% larger value for $f(0, 0)$ of the C_2 Swan bands we will now proceed to establish a value of the solar carbon abundance using this new value. The equivalent width $W_{J''}$ of a molecular absorption transition ($n'', v'', J'' \rightarrow n', v', J'$) is given by (cf. refs. [3, 31])

$$W_{J''} = \frac{\pi e^2}{m_0 c^2} \cdot \lambda^2(v', v'') \cdot f(v', v'') \cdot \frac{S_{J''}}{2J'' + 1} \cdot \int_0^\infty \frac{g(x)}{\kappa} \cdot N_{n'', v'', J''} dx \tag{5}$$

where $N_{n'', v'', J''}$ is the number of absorbing molecules in the initial (n'', v'', J'') state, $S_{J''}$ is the rotational line strength and λ and f have their usual meanings. The integral contains the absorption-emission factor $g(x)$ (which is a function of the optical depth x) and the absorption coefficient κ . If N is given by a Boltzmann distribution, (5) can be written as

$$\log \frac{W_{J''}}{S_{J''}} = \log \frac{W_0}{S_0} - 0.434 \cdot B \cdot \frac{hc}{kT} J''(J'' + 1) \tag{6}$$

where

$$\frac{W_0}{S_0} = \frac{\pi^2}{m_0 c^2} \cdot \lambda^2(v', v'') \cdot f(v', v'') \cdot \int_0^\infty N_{n'', v'', 0} \cdot \frac{g(x)}{\kappa} \cdot dx \tag{7}$$

Here B is the rotational constant for the involved state and T is the absolute temperature. Thus if a number of rotational absorption lines of a given molecular system are observed from a celestial object, a plot of $\log(W_{J''}/S_{J''})$ versus $J''(J'' + 1)$ will give a straight line according to (6). The slope of this line gives the rotational temperature and its intercept with the $\log W_{J''}/S_{J''}$ -axis yields a value for W_0/S_0 from which $N_{n'', v'', 0}$ can be determined according to relation (7).

However, frequently, as in the present case, we want to determine the abundances of the atomic constituents, which build up the molecules N . If we assume that N consists of diatomic molecules $\alpha\beta$ and that the abundances of α and β (relative to hydrogen) are equal to $N(\alpha)$ and $N(\beta)$ eq. (7) can be rewritten in the form

$$\frac{W_0}{S_0} = K \cdot \lambda^2(v', v'') \cdot f(v', v'') \cdot N(\alpha) \cdot N(\beta) \cdot \int_0^\infty \Psi(\alpha, \beta) \Phi(\alpha) \Phi(\beta) \cdot p \times \frac{g(x)}{\kappa} \cdot dx \tag{8}$$

where K is a constant containing a statistical factor and the integral involves (besides the pressure p) the dissociation function $\Psi(\alpha, \beta)$ and the conversion functions $\Phi(\alpha)$ and $\Phi(\beta)$. The function $\Psi(\alpha, \beta)$ contains the dissociation constant D_0^0 for the molecule $\alpha\beta$ and the internal partition functions for α and β . The functions $\Phi(\alpha)$ and $\Phi(\beta)$ convert the number of atoms α and β bound in $\alpha\beta$ -molecules to the total number of atoms α and β (i.e. occurring either as free, neutral or ionized atoms or possibly bound in various molecules).

We now want to determine the solar carbon abundance from observations of the Swan bands observed in the solar photosphere, at first a value of W_0/S_0 have to be determined from eq. (6). From studies of a large number of rotational lines of the

Table III. Some recent determinations of the solar carbon abundance obtained by various methods ($N(H) = 1$, log. scale; $A(H) = 12$).

$N(C) \times 10^4$	$A(C)$	Year	Method
3.55	8.55	1964	Photosp. Swan bands [3]
2.14	8.3	1969	Coronal EUV lines [32]
3.55	8.55	1972	Coronal and chromosp. EUV lines [33]
3.98	8.60	1973	Photosp. forbidden lines [8]
3.55 ± 0.9	8.55 ± 0.10	1973	Recommended average value [4]
2.16 ± 0.12	8.334 ± 0.024	1975	This work, Swan bands, revised f -values

Swan (0, 0) band in the solar spectrum the following value is deduced [3].

$$\log \frac{W_0}{S_0} = 0.29 \pm 0.03 \quad (9)$$

The application of eq. (7) involves the adoption of a given model of the sun's temperature distribution and chemical composition. Adopting the Utrecht Reference Model, $D_0^0 = 6.38$ eV, $\log W_0/S_0 = 0.29 \pm 0.03$ and $f(0, 0) = 0.005 \pm 0.003$ (i.e. the f -value given in ref. [28]) the carbon abundance has been estimated [3] to be

$$N(C) = \left(3.55^{+1.7}_{-1.2} \right) \times 10^{-4} \quad (10)$$

We note that the large error in the determination of the oscillator strength constitutes the main source of the error in $N(C)$ given by (10). We can now easily correct the value (9) using our new f -value and introducing a more realistic D_0^0 value. For the latter purpose we make use of the linear relation between $\log N(C)$ and D_0^0 (for a given f -value) which has been empirically found in the earlier estimates [3] of the Swan bands:

$$2 \log N(C) = -1.05 \cdot D_0^0 - 0.101 \quad (11)$$

This relation was shown to approximate the relation between D_0^0 and $N(C)$ within a few per cent when D_0^0 varied between 3 and 7 eV. Since we have shown the absence of predissociation effects in the $v' = 6$ level of the $d^3\Pi_g$ state this fact, in addition to other evidences, encourages the adoption of the high $D_0^0(C_2)$ values obtained from the earlier mentioned studies of the predissociations in the $C^1\Pi_g$ state as well as recent values obtained by thermochemical methods [2]. Putting $D_0^0 = 6.11 \pm 0.05$ eV the estimate (10) is increased to $N(C) = 4.9 \times 10^{-4}$ according to (11). From eq. (8) we find that (with other parameters remaining constant)

$$N(C) \propto f(0, 0)^{-1/2} \quad (12)$$

Thus introducing the new $f(0, 0)$ value (3) we finally get

$$N(C) = (2.16 \pm 0.12) \times 10^{-4} \quad (13)$$

where the error includes the uncertainties in W_0/S_0 , D_0^0 and $f(0, 0)$. This carbon abundance is 40% lower than the recently "recommended average value" $= 3.55 \times 10^{-4}$ (cf. Table III) which is based mainly upon studies of atomic EUV or forbidden IR lines. Since the f -values applied in the former case are essentially in agreement with results from recent lifetime investigations [9], which are attributed a 10% error, and the theoretical f -values [8] used in the latter case are quoted with a 5% error, we are inclined to believe that the discrepancies originate from differences in the adopted solar models or in the astronomical data. Thus the 40% decrease in the solar abundance demanded from this work calls for closer investigations at this point.

Note added in proof

Another determination of the solar carbon abundance has very recently been published (Mount, G. H. and Linsky, J. L., *Astrophys. J.* 202, L 51 (Nov. 15, 1975)). This determination is based on a non-LTE analysis of CN molecular spectra and yields $A(C) = 8.35 \pm 0.15$ in agreement with the value 8.334 ± 0.024 derived in this work. Thus the 40% decrease of the solar carbon abundance demanded from the present measurements is supported by the new CN work in which it is emphasized that this decrease

has important astronomical consequences. For instance the decreased carbon abundance will affect nucleosynthesis theories and slightly improve the large discrepancy between theory and experiments concerning the solar neutrino flux. It is also pointed out that the new solar carbon abundance will change even the determinations of most stellar carbon abundances which are usually normalized to the solar value.

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