

than the maximum range of the electrons in the material. Consequently only those which happen to have a fairly straight path will emerge and will be counted. Those whose paths are more crooked will not emerge at all and will not enter into the average. Thus there is a selection in favor of the electrons which are least scattered. This probably accounts for the fact that the energy loss for carbon, 0.5 cm, 3 Mev (see Table VI) is about equal to the theoretical, in spite of the fact that diffusion clearly exists. In this case 65 percent of the electrons failed to emerge from the absorber.

After rejecting such a large part of the existing work on energy loss, it seems as though there

remains little upon which to base an opinion as to whether or not theory is in accord with experiment. If, in the three most acceptable measurements on lead, we apply the indicated path length corrections we find that, as an average, the experimental values are at least 1.4 times the theoretical. Aside from the question of whether or not one is inclined to place any faith even in this result, we believe that we have been able to expose some of the pitfalls in energy loss measurement, so that future experiments can be made upon a somewhat firmer basis.

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The Spin of Carbon Thirteen

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Carbon thirteen was concentrated in a Hertz diffusion system to an abundance of 50 percent and enough 35 percent heavy carbon was obtained for a measurement of the nuclear spin of C^{13} . The spin was determined from the relative intensities of the Λ -type doublets of two lines of the O—O Swan band of $C^{13}-C^{13}$. These doublets are so close that a Lummer-Gehrke plate crossed with a 21-ft. grating was needed to obtain sufficient resolving power. An intensity analysis of the combined interference patterns of the two doublets proves that the C^{13} nucleus obeys the Fermi-Dirac statistics and strongly indicates a spin of $\frac{3}{2}$. This is in disagreement with a spin of $\frac{1}{2}$ predicted for this nucleus from the Hartree nuclear model but is the value predicted by the alpha-particle model.

THE determination of the nuclear spin of the ground state of C^{13} has become of considerable interest lately because it is one of the few magnitudes concerning which the predictions from the alpha-particle nuclear model and from the Hartree model are in complete disagreement.¹ The former predicts a spin of $\frac{3}{2}$ and the latter a spin of $\frac{1}{2}$. The most straightforward method of determining the spin is by measuring the relative intensities of alternate lines in the $C^{13}C^{13}$ molecular spectrum. In ordinary carbon, the $C^{13}C^{13}$ bands are only one ten-thousandth as intense as the $C^{12}C^{12}$ bands; so that for a measurement of the C^{13} spin, it is necessary to concentrate the C^{13} isotope considerably.

CONCENTRATION OF CARBON THIRTEEN

The enrichment of C^{13} was carried out in a 34-member Hertz diffusion apparatus described by Wooldridge and Smythe.² To obtain high concentration the diffusion was in two stages. For the first diffusion, pure methane was pumped slowly and continuously through the light end at 1.4 cm pressure to maintain the normal concentration of C^{13} at this end. Six liters of methane at 8 mm pressure and about 7 percent C^{13} could be collected at the heavy end after a five-day run. Due to the large quantity of gas passed through the light end, considerable amounts of heavy impurities also collected in

¹R. G. Sachs, Phys. Rev. 55, 825 (1939).

²D. E. Wooldridge and W. R. Smythe, Phys. Rev. 50, 233 (1936).

the heavy end. The CO and CO₂ were removed continuously by hot I₂O₅ and a liquid-air trap. Nitrogen was removed from the heavy methane by a simple fractionation. Four five-day runs were made to collect enough methane enriched in C¹³ to refill the diffusion system for a final enrichment. The second diffusion yielded a small quantity of methane containing 50 percent C¹³ and about one liter of methane at 1 cm pressure of 35–40 percent C¹³.

SPECTROSCOPIC PROCEDURE

The nuclear spin and statistics may be determined from the band spectrum of a homonuclear molecule by measuring the ratio of intensities of alternate lines, the statistics determining which lines are stronger, and the relative intensities being given by $s/(s+1)$, where s is the spin. Thus for $s=0$, as in C¹², alternate lines are entirely missing, while for spins of $\frac{1}{2}$, 1, and $\frac{3}{2}$ the intensity ratios are $\frac{1}{3}$, $\frac{1}{2}$, and $\frac{3}{5}$, respectively. Unfortunately, the only bands available in the case of carbon are ¹Π–¹Π and ³Π–³Π, so that the lines whose relative intensities must be measured are very close Λ-type doublets. The Deslandres-d'Azambuja (¹Π–¹Π) bands are usually excited by the continuous breakdown of hydrocarbon gases at fairly high pressures in an electrical discharge, and since no method of exciting them which used a reasonably small quantity of carbon could be found, the Swan bands (³Π–³Π) were used. Each member of a single branch in a single band of this system consists of six lines. Near the head there are three well separated lines, each of which is a narrow doublet, the relative intensity of the two doublet components being the desired datum. Far from the head the triplet structure has drawn together into a single line and the doublet structure has spread out so that the lines appear double. In the intermediate region a combination situation holds. The only place where the doublet structure is large enough to resolve and where the desired lines are not interfered with by some overlapping band of C¹³C¹³, C¹²C¹³ or C¹²C¹² seemed to be in the (0–0) band (λ5165) just before it reaches the (1–1) head. Here the triplets of the R branches of C¹³C¹³, C¹²C¹³, and C¹²C¹² lie side by side with little overlapping

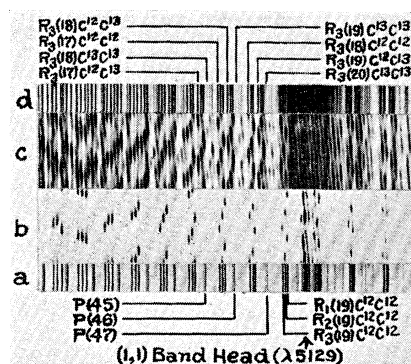


FIG. 1. Spectrogram of carbon bands.

except that one member, R_1 , of each molecule is interfered with by the P branch of the next lighter molecule (see Fig. 1, *a* and *d*). Because the carbon molecule approximates Hund's case "a" coupling for low rotational quantum numbers, the triplet members ³Π₀–³Π₀(R_3) show considerably wider separation of the Λ-type doublets than do the ³Π₁–³Π₁(R_2) and ³Π₂–³Π₂(R_1) the separation of the ³Π₀–³Π₀ lines in question being about 0.14 cm. The resolving power required to resolve these doublets, assuming they are very sharp and have equal intensities is about 140,000. Actually the temperature broadening and inequality of intensity of the two components demanded much higher resolving power.

As no grating of sufficient resolving power was available, a 21-ft. grating was crossed with a Lummer-Gehrke plate having a theoretical resolving power of 300,000. Through the kindness of Dr. Badger of the Chemistry Department an Eagle-mounted concave grating very fast in the second order of the green was used. The somewhat complicated optical system required by the Lummer plate and for correcting the astigmatism of the grating was accomplished by suitable lenses supplied by Dr. Bowen.

Such an optical system requires a powerful light source. A discharge tube with a water-cooled quartz capillary 1 mm in diameter and one cm long was constructed. The heavy methane at about 3 mm pressure was admitted to the tube and a discharge run until most of the carbon was deposited in the tube, principally on the electrodes and in the capillary. The tube was then evacuated and 2 cm of pure argon admitted. When a discharge was run through this tube

from a 20,000-volt, 3-kw, transformer with bad voltage regulation, the Swan bands appeared brilliant for hours. To secure this long life from the small quantities of carbon used, it seemed necessary to exclude mercury vapor and to have the aluminum electrodes polished and with rounded edges.

Several plates of varying exposures were taken with ordinary carbon and with 35 percent C^{13} , and step weakener photographs developed with each plate to make possible accurate intensity measurements. In Fig. 1 *b* and *c* were taken on Eastman I-J plates from ordinary carbon and 35 percent C^{13} with exposures 7 min.–60 min., respectively. The ${}^3\Pi_0-{}^3\Pi_0$ doublets of $C^{12}-C^{13}$, which have equal intensity, are not quite resolved by the Lummer plate, and the corresponding interference fringes of $C^{13}C^{13}$ may be seen to be somewhat asymmetrical, indicating components of unequal intensity.

ANALYSIS OF SPECTRA

There are three lines of measurable doubling, $R_3(17)$, $R_3(18)$ and $R_3(19)$ which, for all the molecules, fall clear of other lines. From the wave-lengths of the C^{12}_2 lines given by Shea,³ the staggering of alternate lines and hence the doublet separation, can be computed roughly. This calculation indicates about 30 percent greater doublet separation for $R_3(17)$ and for $R_3(19)$ than for $R_3(18)$. This was confirmed qualitatively by a visual examination of the fringes and so only the first two were analyzed.

A simple method of determining the statistics obeyed by the C^{13} nucleus is to compare the position of the center of gravity of an R_3 line relative to the corresponding R_3 line in the C^{12}_2 , $C^{12}C^{13}$ and C^{13}_2 bands. Visual measurements of the intensity maxima on a comparator showed at once that the strong component in the C^{13}_2 line was the one which is missing in the C^{12}_2 line, proving that the C^{13} nucleus obeys the Fermi-Dirac statistics. The position of this maximum was visually estimated to be very close to the position of a single component which indicated a very weak second component and a probable spin of $\frac{1}{2}$. Subsequent microphotometer measurements, however, make this

conclusion very doubtful and indicate that it may have been an optical illusion caused by the tendency of the eye to judge density by contrast and thus set the cross hair too near the steep edge of the fringe.

If we are given accurate intensity curves on the same horizontal scale of the $R_3(17)$ or $R_3(19)$ lines of C^{12}_2 , $C^{12}C^{13}$ and C^{13}_2 the most precise analytical procedure is as follows. Two equal C^{12}_2 profiles are drawn with various separations and summed. By adjusting the vertical scales of the resultant curves only one can be made to coincide with the $C^{12}C^{13}$ profile which gives, quite accurately, the doublet separation. With this separation we now draw two C^{12}_2 curves whose intensities are in the ratio $\frac{1}{3}$, $\frac{1}{2}$, $\frac{2}{3}$, etc., and add them. The vertical scale of the resultant curves are adjusted to fit as closely as possible the C^{13}_2 profile. Only one can be made to fit and this gives the ratio of intensities and hence the spin.

To obtain these intensity curves two plates were available, one being about three times as heavily exposed as the other. On both plates there was a uniform change in intensity across the plate, the intensity ratio of the outside orders being about 2 to 1. On the lighter plate eight or nine orders of each line of C^{12}_2 and $C^{12}C^{13}$ were of suitable density for microphotometering. On the heavier plate eight orders each of the C^{13}_2 lines were suitable but only from 3 to 6 each of the C^{12}_2 and $C^{12}C^{13}$ lines, the others being too dense. Microphotometer curves were run on all orders of the lines involved on both plates, the vertical height averaging about 2.5 cm and the width from 2.5 to 5 cm. From the appropriate step weakener picture appearing on each plate the line profile was converted, point by point, into an intensity profile. The resultant curves present a sawtooth appearance due to the grain of the plate, the average grain size being about $1/15$ the width of the line. To determine how completely this grain would average out a microphotometer curve on the same scale was run along one step of the step weakener picture and several such traces were superimposed at random and averaged point by point. The average of eight such curves deviated from a straight line by a maximum of 1 mm in a 3-cm length and crossed a ruled line 20 times in this distance, indicating that the average of eight orders

³ J. D. Shea, Phys. Rev. **30**, 825 (1927).

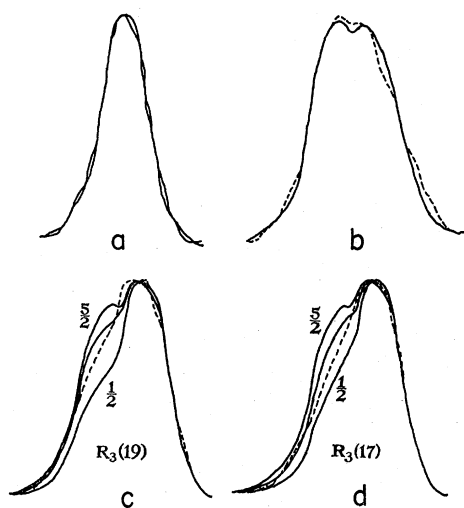


FIG. 2. Microphotometer curves of carbon lines.

should eliminate the grain structure almost completely. This was borne out by the fact that all average curves appeared smooth without artificial smoothing except such as inadvertently occurred by being unable to follow microscopic irregularities through the tracing paper.

The unsmoothed means of 7 orders of $R_3(18)$ C^{12}_2 and 8 orders of $R_3(17)$ C^{12}_2 from the light plate are shown superimposed in Fig. 2a. It is evident that these two lines have the same shape and we will therefore be justified in using the mean of the same lines on the heavy plate to find the shape of the line to be used in analyzing the C^{13}_2 lines on that plate. From two curves of this shape with a separation of 1.30 cm we obtain the solid curve, which is shown in Fig. 2b, superimposed on the observed dotted curve which is the mean of six orders of $R_3(19)$ $C^{12}C^{13}$ from the light plate. A 1.25-cm separation fits $R_3(17)$ $C^{12}C^{13}$ better on the sides but not as well on the top.

The same shape for a single line cannot be assumed for the heavily exposed plate since

changes of temperature of the Lummer plate might have produced a shift in position during exposure. A mean of 3 orders of $R_3(17)$ and four orders of $R_3(18)$ was therefore used for the standard profile on this plate. This turned out to coincide exactly with that of the light plate. The doublet separation was checked on the 3 orders of $R_3(17)$ $C^{12}C^{13}$ and 4 orders of $R_3(19)$ $C^{12}C^{13}$ available on this plate and gave 1.20 and 1.25, respectively, the irregularities being noticeably greater near the tops of the curves as might be expected from the smaller number averaged. The shapes of the peaks to be expected on this plate for $R(19)$ C^{13}_2 with spins of $\frac{1}{2}$, $\frac{3}{2}$ and $\frac{5}{2}$ were then constructed by adding peaks of the standard line shape spaced 1.30 cm apart with intensity ratios 1 to 3, 3 to 5 and 5 to 7, respectively. The resultant curves with the vertical scale suitably adjusted are shown superimposed on the observed dotted mean of 8 orders of $R_3(19)$ C^{13}_2 in Fig. 2c. Corresponding curves for $R_3(17)$, using a 1.25-cm spacing, are shown in Fig. 2d. All curves fit almost exactly for two-thirds of the distance up the steep side. On the side of the weak component the $\frac{3}{2}$ curve fits well on the average but the bump which should be present is absent. No known smoothing process, such as the finite microphotometer slit width, could eliminate this bump completely. Accidental deviations of this order have been observed as shown in Fig. 2b, but would be unlikely to occur in both lines. On the other hand, the observed curve is much too wide to agree with the calculated $\frac{1}{2}$ curve. No accidental width deviations of this size have been observed even between single unaveraged curves. The standard shape used for the single line was well determined so the use of a wider standard cannot be justified. Use of a wider separation improves the $\frac{1}{2}$ fit at the base but makes it worse in the middle. The spin of 1 is excluded by the statistics.

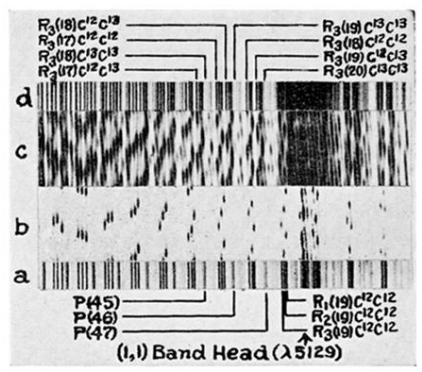


FIG. 1. Spectrogram of carbon bands.