Beta Spectra of C¹⁴ and S³⁵

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The beta spectrum of carbon 14 was measured with radioactive methane as the source in a proportional counter. To reduce end and side wall effects a pressure of 6 atmos (argon) was employed and a magnetic field of 3500 gauss was applied parallel to the counter axis. The spectrum of sulfur 35 was observed with the same equipment. The theoretical allowed spectrum for this source was modified according to calculations made to take into account the instrumental limitations. The modified theoretical form agreed excellently with the observations down to energies below 5 kev. With the sulphur spectrum as an experimental standard for an allowed shape very accurate empirical correction factors could be obtained, and these were applied to the data for C14. The adjusted data for C14 were found to follow the theoretical shape for an allowed transition to within 1 percent accuracy at energies above about 3 kev.

INTRODUCTION

HE beta decay of C¹⁴ has been investigated by many,¹ but some unsatisfactory features remain. The spins of both C¹⁴ and N¹⁴ have been measured directly, giving I=0 and I=1, respectively. A spin change of unity is involved in the transition which is allowed, unfavored, according to Gamow-Teller rules. The observed comparative half-life ft is 1.9×10^9 sec, which exceeds the average value by a factor of about 104, and suggests empirical classification among second forbidden. Feenberg² suggested that an explanation of the anomaly was possible on the basis of a transition from an S state in C^{14} to a pure D state of N^{14} where LS coupling prevails. A small admixture of ³S, less than 10^{-2} , to account for the large ft value may dominate the beta decay and produce a close approximation to an allowed shape. The shape of the spectrum is of enhanced interest in this case. There is general agreement that the shape of the spectrum is allowed down to about 50 kev, but below that there is some disagreement. Wu³ found the Fermi plot linear down to 25 kev. Recently Mize and Zaffarano,⁴ with a thin source in a proportional counter, found that the intensity dropped below the theoretical value at energies below about 50 kev. Their method seemed to work correctly with S³⁵ and Pm¹⁴⁶.

Most of the experimental difficulties in the lowenergy region arise in the use of solid sources and here the proportional counter with a gaseous source can be applied most advantageously. In fact, C¹⁴ has been investigated in this way by Angus, Cockroft, and Curran⁵ and their results showed a deficiency of lowenergy particles. Since then the technique has improved very considerably. Thus field-correcting tubes have been introduced,⁵ thereby removing much of the uncertainty associated with the elimination or control of end effect by other means. The use of strong axial magnetic fields to reduce wall effect has proved valuable. For these reasons it was clear that an accurate examination of the C¹⁴ spectrum with the improved proportional tube spectrometer was desirable.

CHARACTERISTICS OF THE SPECTROMETER

With field tubes at the correct potential the gas multiplication factor is effectively constant throughout the volume of the cylinder of length equal to that of the operating central wire. With radioactive gas as a constituent of the filling mixture the beta spectrum can be analyzed successfully, provided the quantity of such gas does not adversely affect resolution. A strictly accurate spectrum is observed, provided each beta particle spends the whole of its energy in the sensitive volume and no electrons enter from outside. This condition can be satisfied at best approximately at high energies. From this point of view both S³⁵ and C¹⁴ must be regarded as intermediate cases (end points at 167 and 156 kev, respectively).

Suppose that ρ nuclei decay per second and per cm³ and that the fraction of beta rays with energy between E and E+dE is N(E)dE. In the sensitive volume V we have $N\rho V dE$ such electrons produced per second. If they expend their energy within the volume V they give rise to pulses proportional in amplitude to the energy E and they are counted with an analyzer as occupying the channel E of width dE. Some of them escape from the sensitive volume or they reach the cylinder and so give rise to smaller pulses which are counted in lower-energy channels. If R is the particle range corresponding to the energy E it can be shown that for the electrons $N\rho R/4$ escape per cm² per sec through the surface S of the sensitive volume. Thus, $N\rho V dE [1 - \frac{1}{4}R(S/V)] = N\rho V dE (1 - \delta_E)$ are counted correctly in channel E. Here δ_E is the fractional loss to lower channels. At both ends of the cylindrical counting volume V, the rate of entry into V of electrons of energy E is $N\rho R/4$ per cm². Again these electrons spend part of their energy in V and give rise to smaller

¹Hollander, Perlman, Seaborg, Revs. Modern Phys. 25, 478 (1953).

^{(1953).}
² E. Feenberg and K. C. Hammack, Phys. Rev. 75, 1877 (1949).
³ L. Feldman and C. S. Wu, Phys. Rev. 75, 1286 (1949).
⁴ Summary Report of the Indiana Conference on Nuclear Spectroscopy and the Shell Model, May, 1953 (unpublished).
⁵ Angus, Cockroft, and Curran, Phil. Mag. 40, 522 (1949).
⁶ A. L. Cockroft and S. C. Curran, Rev. Sci. Instr. 22, 37 (1951).

pulses. If the specific ionization of electrons can be assumed constant, pulses caused by electrons escaping from or entering the sensitive volume are spread uniformly into all channels below E. In the low-energy region this is too approximate an assumption, and the spreading has to be determined by numerical integration, applying the range-energy relationship.

For a cylindrical counter of radius r and length l, the fraction δ_E not counted correctly in channel E is, as shown above,

$\delta_E = R/2l + R/2r,$

so that r and l should be as large as practicable, while the range R of the beta rays should be small. A gas of high atomic number helps to restrict R but xenon is difficult to procure in adequate quantities. Here argon at pressures up to 7 atmos was used. The dimensions were r=6.8 cm, l=19.5 cm. At the pressure of 5.5 atmos of argon, frequently employed, the range of electrons of energy 100 kev is about 2 cm, not small compared with r. But the application of an axial magnetic field B, 3500 gauss, forces the primary particles to describe circles of radius $r_m = mv/eB$ and as the particle loses velocity v the radius r_m rapidly decreases. The wall effect is so reduced at least in the ratio $R/2r_m$. Table I shows some calculated corrections. Separate experiments showed that the end effect was reduced by the field to approximately half the value in the absence of field. This was ascribed to the influence of the field on the number leaving and entering the ends.

The fraction δ_E of electrons in channel E which give smaller pulses to the channels below E decreases with energy and is less than 2 percent below 25 key, while that caused by more energetic electrons increases. The magnetic field effect makes the calculations rather approximate, but the results are satisfactory, since the correction refers to something like 5 percent of the particles. The nature of the particle trajectories, which are particularly irregular at low energies as a result of scattering, adds to the uncertainties. The corrections as applied to S35 yield very satisfactory agreement between the theoretical and experimental spectra, as is shown below. This allows us to proceed in an alternative manner with the study of C14. The spectra of C14 and S³⁵ have similar limiting energies and the latter has been shown, and is shown here, with the application of the corrections, to have a very straight Fermi plot down to energies below 5 kev. Taking the experimental

TABLE I. Correction factors at 5.5 atmos pressure and 3500 gauss.

Electron energy in kev	R/21 %	r _m /r %	
25	0.6	2.2	
51	1.7	3.3	
102	5.7	4.6	
153	11.0	5.7	



FIG. 1. Curves A_1 and B_1 : observed spectra of S³⁵ and C¹⁴ respectively; Curves A_2 and B_2 : corresponding theoretical spectra, assuming the transitions allowed.

results for S³⁵ as standard for an allowed source, we can deduce by comparison with the theoretical spectrum the precise correction to apply at any particular value of energy. In this way the observations for a spectrum such as that of C¹⁴ can be corrected accurately. The corrections throughout the range are relatively small and the corrected experimental data can be compared closely with the theoretical value. It is estimated that the true intensity is found to better than 1 percent accuracy in this manner. This technique is applied to C¹⁴ in Table I. It is worth stressing that this comparative method of study of beta spectra is one of fairly wide applicability. A number of accurately studied spectra, such as that of S³⁵, are available, and any instrument subject to errors and uncertainties as an absolute tool can nevertheless prove reliable in relative work. This is particularly true for the proportional counter as completely equivalent sources, as regards thickness, etc., may be difficult to make in solid form, but are readily produced as gaseous constituents of the gas mixture used in the counter.

EXPERIMENTAL ARRANGEMENT

The choice of a suitable gas for the analysis of C¹⁴ was determined solely on considerations of resolving power. Here methane suggested itself as eminently suitable. Carbon dioxide was liberated from barium carbonate by vacuum heating and converted to methane by adding hydrogen. Ruthenium was used as a catalyst; it was placed in a Pyrex tube and heated to 250°C. About 10 forced passages of the gases over the catalyst completed the reaction $(CO_2+4H_2\rightarrow CH_4+2H_2O)$. Excess hydrogen was removed through a heated



FIG. 2. The theoretical spectrum of S³⁵, shown by the filled circles, is modified, according to calculations of the instrumental effects, to the shape shown by the full curve. The open circles show the experimental results, indicating close agreement.

palladium tube. The radiomethane required in the counter was small in amount (<0.01 percent) and the main mixture consisted of methane to a pressure of 10 cm Hg and argon to a pressure of 6 atmos. Calcium turnings were used in the attached purifier. The width at half-height of the K_{α} line of silver was 15 percent. For the work on S³⁵ labeled CS₂ vapor was used. Neither radioactive source reacted with the counter walls. The voltage on the counter was about 6500 volts. A nickel wire delay line kicksorter of the Hutchinson-Scarrott⁷ type was used. This 100-channel pulse analyzer accepts pulses from 4.1 volts to 49 volts in amplitude. In this work it was mostly used on the 34-channel setting. Statistical errors of less than 2 percent could be achieved in a run lasting a few minutes. The kicksorter channels were calibrated in volts using the builtin pulse generator and calibrated attenuator. In energy units the calibration was effected with the fluorescence x-rays of silver. The counting rate was adjusted to about 14 000 per min. The background rate was about 450 per min. The complete spectra and various parts were analyzed many times and the results were self-consistent. The theoretical spectra were calculated using the Bethe-Bacher approximation for the Coulomb (Fermi) factor.

RESULTS AND DISCUSSION

In Fig. 1, one set of measurements for S^{35} and C^{14} are shown. In the upper figure the theoretical spectra for both are given. It is clear that the experimental results differ from the theoretical in the same way in each case. There is obviously about the same amount of distortion produced at low energies where some excess of particles caused by the end and wall effects already discussed are apparent. Both the calculated and experi-

mental spectra crossover in the same energy channel. It is obvious that the distortion is about the same in amount for each spectrum and we can say that if S^{35} is allowed in form the experimental data for C^{14} is such as to indicate, to a fairly close approximation, that C^{14} is allowed in form.

For more detailed comparison of theory and experiment graphical calculation of the correction per channel was made for S³⁵ and C¹⁴. The results of this treatment, along the lines discussed above, are given in Fig. 2 for S³⁵ only. The results for C¹⁴ not reproduced here took a very similar form, but we prefer to apply the alternative method of correction to this source as indicated below. In making the graphical calculations the spectrum was plotted in terms of range R rather than energy E, using the range-energy relationship for argon. It should be remarked that the total fraction of electrons entering the sensitive volume if no magnetic field is applied is $[(1/2l) \int_0^R NR dR] / \int_0^R NdR$, which in the present case is 3 percent. If magnetic field is applied this fraction becomes 1.6 percent as shown above. This must be doubled to 3.2 percent, since an equal percentage of the detected electrons pass out of the volume. The wall effect falls from about 9 percent (expressed in the same way) to about 2 percent in the case of field applied. Hence the total of wrongly analyzed pulses amounts to approximately 5 percent. This total agrees well with the sum of those spread into lower channels. The validity of the correction procedure was checked at different values of gas pressure and magnetic field strength.

It can be seen from Fig. 2 that S^{35} yields results entirely consistent with an allowed type of decay. This gives, in conjunction with earlier results, complete confidence in adopting S^{35} as a standard allowed spectrum for comparison with that of C^{14} .

To compare the results for C^{14} with theory the experimental curve for S^{35} was superimposed on the theoretical curve for the same source (curves A_1 and



FIG. 3. The full curve represents the theoretical spectrum of C¹⁴, assumed allowed. The circles show the corrected <code>_experimental data</code>.

⁷G. W. Hutchinson and G. G. Scarrott, Phil. Mag. 42, 792 (1951).

 A_2 of Fig. 1) normalizing for the same number of particles. In this way an accurate correction factor for beta intensity observed at any particular energy was deduced. This factor was then applied to the experimental data for C^{14} (curve B_1 of Fig. 1), and in Fig. 3 we show separately a plot of this corrected data. The full curve in Fig. 3 represents the theoretical form of the C^{14} spectrum, if the decay is allowed. It is seen to fit excellently the adjusted experimental values and the

agreement from less than 3 kev upwards constitutes strong evidence that the carbon spectrum is allowed in form.

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Total Cross Sections of 208-Mev and 315-Mev Protons for Light Elements*

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Transmission measurements were performed with an external collimated proton beam of The University of Chicago 170-inch synchrocyclotron. Total cross sections of the neutron and ten light elements (H and D, Li, B, Be, C, N, O, Al, S, and Cl) were measured by coincidence and anticoincidence methods at proton energies of 208 and 315 Mev at several subtended angles.

The cross sections obtained include all processes except the Coulomb scattering, for which corrections were applied. The corrected cross sections, measured at different subtended angles ("poor geometries") were plotted with their statistical uncertainties as a function of the solid angle subtended or of the angle, and extrapolated to zero degrees ("good geometry").

Only a limited number of previous investigations of the light elements at high particle energies have been made. The results found in the present study for the proton total cross sections for the various elements were comparable to the available published neutron total cross sections measured at the same energies.

It was found, by application of the transparent optical model, that cross sections measured were consistent with a nuclear radius $R = 1.23A^{\frac{1}{3}} \times 10^{-13}$ cm and an absorption coefficient $K = 0.5 \times 10^{13}$ cm⁻¹.

I. INTRODUCTION

HE measurement of the total cross section of nuclei for high-energy nucleons gives information about the nuclear radius and the behavior of the highenergy nucleons within nuclear matter. In particular it gives information about the Serber¹ transparency effect of nuclear matter to high-energy nucleons which up to the present time has been studied mainly with neutrons.

In the past most of the total cross-section measurements were carried out using neutrons because of the difficulties involved in experiments with protons. The main results of this work with neutrons are well summarized in a recent paper by Nedzel.²

In principle, the measurement of total cross sections for neutrons does not present a complex problem. Neutrons have no charge and consequently their electromagnetic interactions are extremely weak. Therefore, nuclear forces cause by far the most important interaction between high-energy neutrons and nuclei.

The measurements of neutron total cross sections are not complicated by the large contribution of the nuclear Coulomb field, which in the case of proton scattering is responsible for a fast rise of cross section at small angles.

In general one expects that the cross section depends on the energy. Since as a rule high-energy neutrons are not monoenergetic it is useful to calculate an effective neutron energy \bar{E}_{eff} . This is an involved problem, which is a serious handicap for the interpretation of the measurements. A second disadvantage follows from the fact that high-energy neutrons are detected indirectly by nuclear reactions or by recoil protons produced in neutron-proton scattering.

In contrast to the neutron situation, collimated monoenergetic beams of protons are readily obtained at high energies. Owing to energy loss by ionization, however, protons of several-hundred Mev energy are not able to cross even one entire nuclear interaction mean free path. In order to measure the nuclear interaction at a given energy it consequently becomes necessary to use relatively thin absorbers. As another difficulty, Coulomb scattering does not allow the direct "good geometry" measurements of total nuclear cross sections, but these can be obtained indirectly by a

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¹ R. Serber, Phys. Rev. **72**, 1114 (1947). ² V. A. Nedzel, Phys. Rev. **94**, 175 (1954).