Extension of the Radioactive Series, $Z=N\pm1$

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(Received July 29, 1941)

The radioactive positron emitters Si^{27}, S^{31} and A^{35} were produced by 15-Mev alpha-particles while Sc^{41} was obtained by 8-Mev deutrons. The half-lives of these elements have been determined as 4.92 ± 0.1 seconds, 3.18 ± 0.04 seconds, 1.88 ± 0.04 seconds, and 0.87 ± 0.03 seconds, respectively. The upper limit of the energies of the positrons emitted by S^{31}, A^{35}, and Sc^{41} were measured and found to be 3.87 ± 0.15 Mev, 4.41 ± 0.09 Mev, and 4.94 ± 0.07 Mev, respectively. The Coulomb energies of the "extra proton" of the nuclei of the $Z-N=1$ were calculated according to two nuclear models. The variations in the Coulomb energies indicate a nuclear "shell" structure similar to that given by the Hartree model of the nucleus.

RADIOACTIVE nuclei for which $Z-N=\pm1$ are of particular interest in nuclear physics. It is possible to deduce the difference of Coulomb energy of an isobaric pair of nuclei of this type from the measured mass difference and from these measurements the radius of each of these nuclei can be calculated.[1,2] The high degree of symmetry of the nuclei of this type enables one to determine theoretically the difference in Coulomb energy. The calculation depends upon the assumption that the binding energy due to the specific nuclear forces is the same for each member of a pair of these isobaric nuclei.[1,2] Taking into account the conservation of energy it follows that the Coulomb energy associated with the "extra" proton is given by

$$E_c=E_{\max}+1.78 \text{ Mev}, \qquad (1)$$

where $E_{\max}$ is the upper limit of the positron spectrum.

The shortest periods of the series $Z-N=+1$ have been investigated in several laboratories[3-5] by different methods. The measurement of very short periods presents two difficulties not ordinarily encountered. The time lag of the recording instrument should be small compared to the period being measured, and the time intervals used should be equal in length or short compared to the half-life. In this laboratory a Geiger counter scaling circuit was used. The time lag of such a circuit is less than 10^{-4} second, and the sensitivity extends over a wide range, which makes it possible to use equal time intervals for a short period and still make proper corrections for the background. With a scale-of-16 it was possible to obtain accurate counting rates of 50,000 counts per minute. For the shortest periods measured it was necessary to use time intervals of about one-half second. This means that only 400 counts, at most, would appear in each time interval. Since the probable error of so few counts is of the order of five percent it is seen that such data give rather large statistical fluctuations.

In order to improve the statistics of these data the most obvious method is to increase the number of counts occurring in any time interval. Since this cannot be done directly because of the limited speed of the mechanical recorder, a procedure was followed that gives the same effect indirectly.

Each activity curve was measured several times (say n) for equal time intervals and the total number of counts occurring in the n first intervals, the n second intervals, etc., were plotted. Then the resulting "sum" curve is an activity curve in which the effective number of counts per time interval will be increased approximately n times that of any one set of data, and thus the statistical fluctuations will be reduced. This method made it possible to obtain at least 2000 counts for every point used in the determination of the half-lives. The samples do not have to be of the same strength, since no

[1] E. Wigner, Phys. Rev. **56**, 519 (1939).
[2] H. A. Bethe, Phys. Rev. **54**, 436 (1938).
[3] G. Kuerti and S. N. Van Voorhis, Phys. Rev. **56**, 614 (1939).
[4] White, Delsasso, Fox, and Creutz, Phys. Rev. **56**, 512 (1939).
[5] L. D. P. King and D. R. Elliott, Phys. Rev. **58**, 846 (1940).

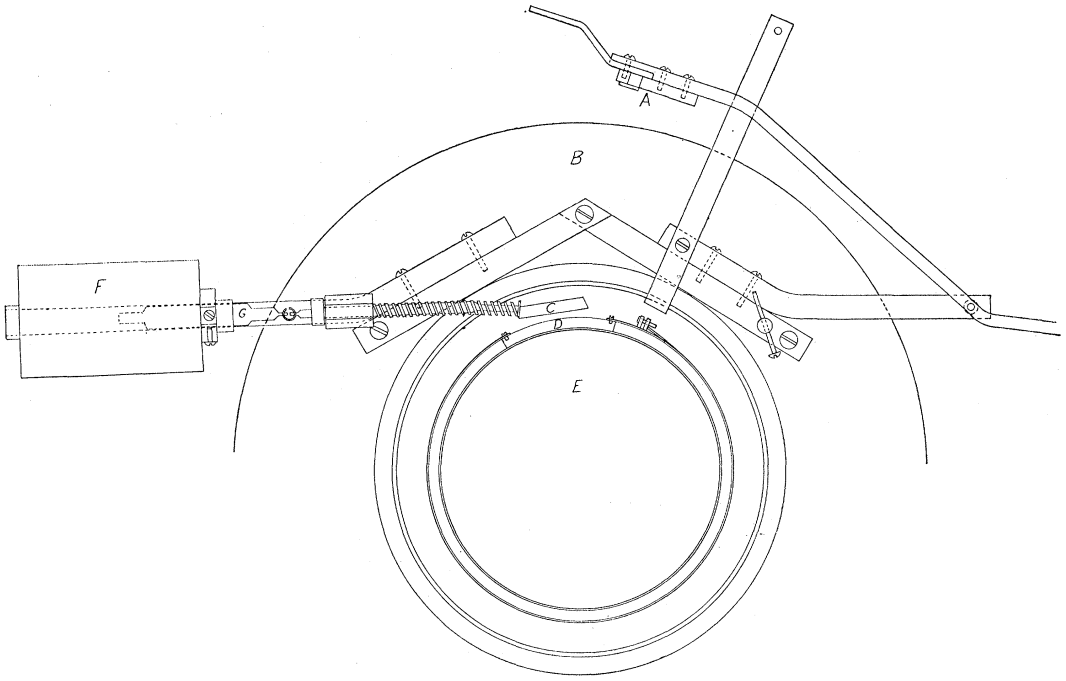

FIG. 1. Target holder of cloud chamber. The target holder *A* moves into place just behind the shutter *C*. *A*—target holder; *B*—coil face; *C*—shutter; *D*—window; *E*—chamber; *F*—solenoid; *G*—core.

error arises due to variations in beam energy, bombardment time, etc., if one is interested only in the rate of decay.

This procedure may be justified in the following way. In the first place, it is certainly equivalent to measuring the activity curves of all n samples simultaneously with n counters and recording only the total number of counts occurring in each time interval. This, again, is equivalent to measuring the activity curves of one sample consisting of the sum of the n samples with a single counter capable of registering the total number of counts.

The dial readings of the mechanical counter and a stop-watch were photographed by an electrically driven camera using a shutter speed of 1/100 second. Accurate timing was obtained by a motor-driven cam switch, the variations in speed of which varied less than 0.1 percent. The stop-watch readings served only as a check. After the short-lived activity had disappeared, the background reading was taken so that proper corrections for any long period components of the activity curve could be made.

The short half-lives of these elements made it necessary to bombard the samples outside of the cyclotron. The beam was brought into the air through a 0.006-inch aluminum window. The time required to transfer the samples from the beam to the recorder was less than 2 seconds. In order to eliminate any background due to air recoil nuclei, the samples were bathed in a hydrogen atmosphere during bombardment. In both the period and energy measurements several samples were used in rotation in order to reduce the influence of any longer periods. The cyclotron was "on" only during actual bombardments which varied from $\frac{1}{2}$ to 5 seconds depending on the sample used. Beam intensities of $\frac{1}{20}$ microampere were used in both the alpha- and deuteron bombardments.

Energy measurements were made by means of a cloud chamber. Figure 1 shows the arrangement of the source relative to the chamber. The positrons emitted from the source pass through a copper window $1\frac{1}{2}''$ long by $\frac{1}{2}''$ high, and 0.001″ thick located in the wall of the chamber. Since tracks entering the chamber before expansion may appear diffuse, a shutter made of $\frac{1}{4}''$ brass was placed between the window and source. The

shutter was drawn back immediately after the chamber expanded. By placing the source about two inches in front of the window, a partial collimation was obtained. With this arrangement a very high percentage of the tracks traversed the full width of the chamber, thus making their measurement both easy and accurate. Moreover, it is seen that with proper magnetic field adjustment one-third of the chamber is accessible only to the highest energy rays. Thus it was possible to use a strong source and favor a high energy end of the spectrum, thereby reducing to a minimum the number of exposures needed to determine the upper spectrum limit. An average of four to five measurable tracks was obtained in each exposure. Because of the high energies of the positrons measured, an air-filled chamber was satisfactory. Only one photograph was made for each bombardment and four bombardments were made per minute. The radii of the tracks were measured by reprojecting them onto a screen by the same optical system used in taking the pictures. The curvatures of the tracks were then measured by comparing them with arcs of circles of known radii ruled on a thin celluloid sheet.

$_{14}Si^{27}$

The Rochester group,[3] and more recently the Princeton group,[4] have obtained this radioactive isotope by using high energy protons on aluminum. In this laboratory[5] Si^{27} was produced by alpha-bombardment of magnesium, Mg^{24} (α, n) Si^{27}. A 15-Mev alpha-beam from the Purdue cyclotron was used. Magnesium has three stable isotopes of mass numbers 24, 25, 26 with abundances of about 77, 12, 11 percent, respectively. Alpha-bombardment of magnesium 25, and 26 gives rise to the activities of 2.4 minutes and 6.7 minutes, respectively,[6] which form the background of the activity curve shown in Fig. 2. Analysis of this curve yields a value of 4.92 ± 0.1 sec. for the half-life of Si^{27} in agreement with the Princeton value of 4.9 ± 0.2 seconds.

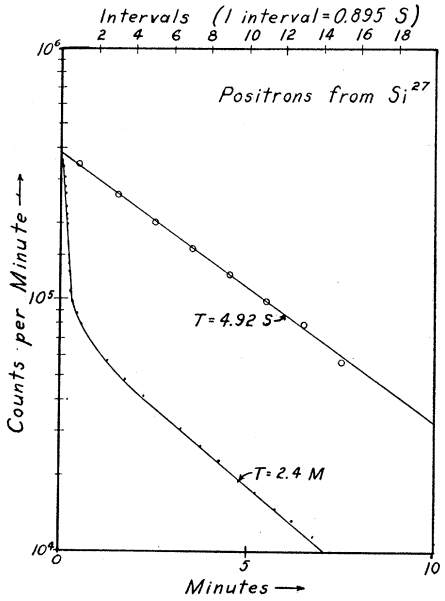

FIG. 2. Decay curve of S^{27} produced by $_{12}Mg^{24}$ (α, n) $_{14}Si^{27}$.

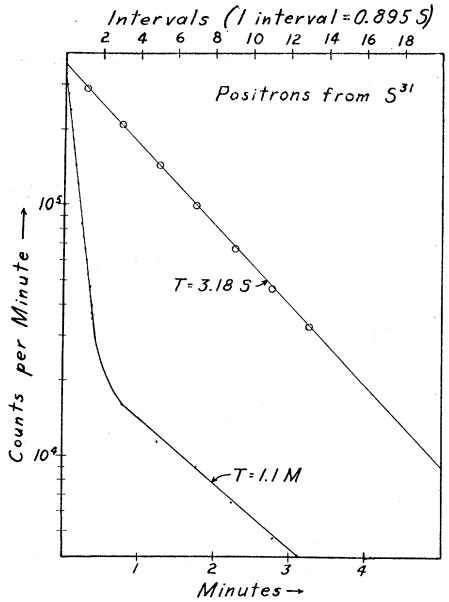

FIG. 3. Decay curve of S^{31} produced by $_{14}Si^{28}$ (α, n) $_{16}S^{31}$.

No energy measurements were made for Si^{27} in this laboratory.

$_{16}S^{31}$

S^{31} was produced by alpha-bombardment of Si in the form of quartz according to the reaction Si^{28} (α, n) S^{31}.[5] Quartz was used because of its high purity. The oxygen in the quartz causes no difficulty, for bombardment of oxygen by a 15-Mev alpha-beam produces no activity. No attempt was made to eliminate the air recoils in this case, thus accounting for the 1.1-minute

[6] H. A. Bethe and W. J. Henderson, Phys. Rev. **56**, 1060 (1939).

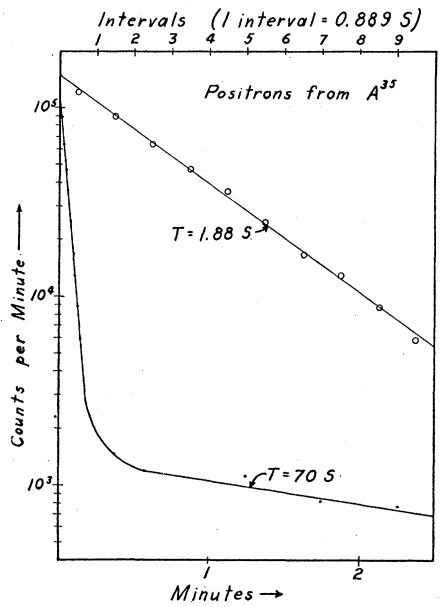

FIG. 4. Decay curve of S^{35} produced by $_{16}S^{32}$ (α, n) $_{18}A^{35}$.

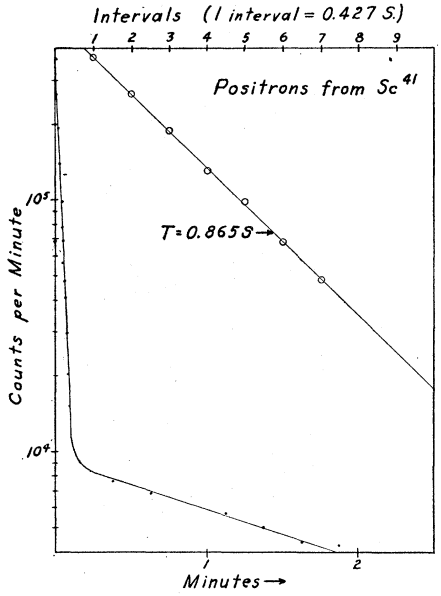

FIG. 5. Decay curve of Sc^{41} produced by $_{20}Ca^{40}$ (d, n) Sc^{41}.

period shown in Fig. 3. The assignment of the 3.18 ± 0.04-second period to S^{31} seems quite certain. In the first place, all other nuclei from probable alpha-reactions have been assigned with certainty. Also the Rochester group (private communication) and the Princeton group[7] have obtained the same half-life (3.2 ± 0.2 seconds) by proton bombardment of P^{31}.

The Princeton group report a value of 3.85 ± 0.07 Mev for the upper limit of the positron spectrum. This is to be compared with our value of 3.87 ± 0.15 Mev.[8] Both measurements were obtained by cloud-chamber methods.

$_{18}A^{35}$

A^{35} was obtained by the bombardment of sulphur with 15-Mev alphas.[5,8] Sulphur samples were prepared by melting flowers of sulphur by gentle heat until a thin straw-colored liquid was obtained, and then pouring it into a clean iron mold and allowing it to set. The value of 1.9 ± 0.1 seconds was obtained for the half-life. Because this was in disagreement with the value of 2.2 ± 0.2 sec. published later by the Princeton group,[7] the measurement of the half-life was repeated and a value of 1.88 ± 0.04 seconds was obtained (see Fig. 4). This time a filter of $\frac{1}{8}''$ Al was placed between the active sample and the counter, in order to reduce the low energy com-

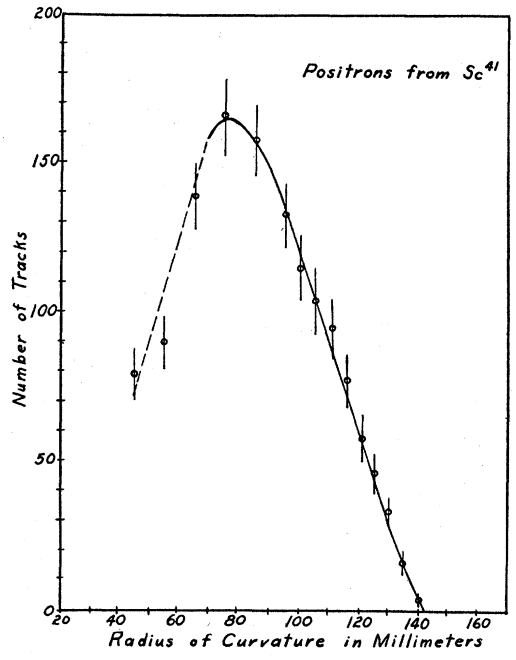

FIG. 6. Positrons from Sc^{41}. Maximum energy 4.94 ± 0.07 Mev, half-life 0.87 ± 0.03 second.

[7] White, Creutz, Delsasso, and Wilson, Phys. Rev. **59**, 63 (1941).

[8] D. R. Elliott and L. D. P. King, Phys. Rev. **59**, 403 (1941).

ponents of the background. The background correction for the resulting activity curve changed the initial slope by less than two percent.

The value of 4.38±0.07 Mev obtained by the Princeton group for the energy of the upper limit of the positron spectrum of A^{35} compares well with our value of 4.41±0.09 Mev.[8]

$_{21}Sc^{41}$

The production of Sc^{41} was obtained by deuteron bombardment of calcium according to the reaction Ca^{40} (d, n) Sc^{41}. The samples used consisted of calcium metal turnings waxed to small brass supports, so that no part of the support was exposed to the deuteron beam. Measurement of the half-life yielded a value of 0.87±0.03 second. (See Fig. 5.) A $\frac{1}{8}''$ aluminum filter placed between the active sample and the counter suppressed all other periods so that their combined effect was reduced from twenty-five percent to less than two percent of the initial activity. Because Sc^{41} cannot be reached by any other of the usual nuclear reactions its identification is mostly one of elimination. All other probable Ca $(d, -)$ reactions have been investigated carefully.[9] Further identification of the 0.87-second period with Sc^{41} is given also by the fact that both the value of the period and the value of the upper energy limit of the positrons are in accord with the values obtained for these quantities by extrapolation from other members of the series $Z-1=\pm 1$ (see Figs. 7, 8, and 9).

Energy measurements made by a cloud chamber gave a value of 4.94±0.07 Mev[8] for the upper energy limit of the positron spectrum (see Fig. 6). The unusually high value of the positron energy eliminates the possibility that impurities are effecting the value of the upper energy limit. The complete absence of a "tail" may indicate that, in many cases, the presence of a "tail" in a distribution curve is caused by an impurity. Because of the changing geometry of the experimental set-up for different energy particles and the increased chance of stray tracks at lower energies it would be difficult to compare the entire curve with the theoretical Fermi curve even if an attempt had been made to count all

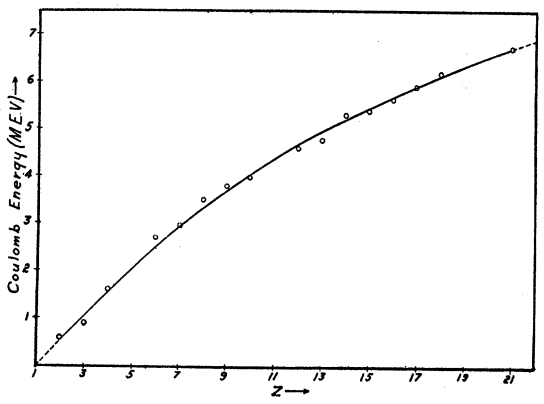

FIG. 7. Experimental values of the Coulomb energy of the "last" proton calculated by Eq. (1) for nuclei with one more proton than neutron, plotted as a function of atomic number. $E_c=E_{max}+1.78$.

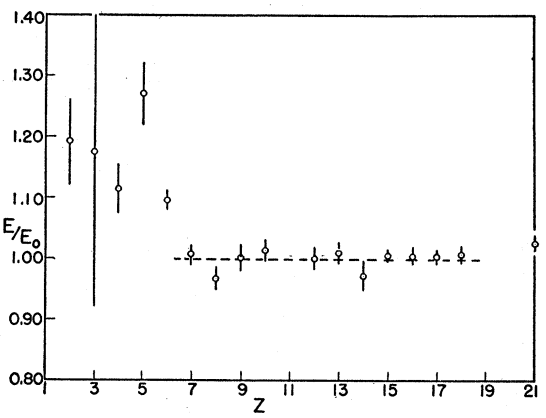

FIG. 8. Ratio of the calculated Coulomb energy to the experimental values. $E=0.592(A-1)/A^{\frac{1}{3}}$.

TABLE I. *Calculation of Coulomb energies, "core" model.*

Z	ϵ	b	ρ	R_0	$f(\rho)$	E (MEV)
2	6.65	1.118	0.618	1.80	0.289	0.70
4	7.65	0.92	0.374	2.46	0.324	1.70
5	1.85	1.84	0.689	2.67	0.280	1.80
6	12.35	0.704	0.244	2.90	0.347	2.57
7	5.3	1.065	0.348	3.06	0.328	2.77
8	11.6	0.715	0.222	3.22	0.352	3.30
9	4.5	0.145	0.341	3.35	0.330	3.40
10	11.0	0.73	0.210	3.49	0.355	3.95
11	8.0	0.852	0.236	3.61	0.350	4.18
12	13.0	0.669	0.180	3.73	0.361	4.59
13	7.5	0.877	0.228	3.84	0.351	4.72
14	11.9	0.696	0.117	3.94	0.362	5.14
15	8.9	0.803	0.191	4.03	0.359	5.38
16	12.9	0.667	0.162	4.12	0.365	5.63
17	9.6	0.772	0.183	4.22	0.361	5.89
18	9.6	0.772	0.180	4.30	0.361	6.14
21	10.0	0.75	0.166	4.53	0.364	6.92

[9] G. T. Seaborg, Chem. Rev. **27**, 199 (1940).

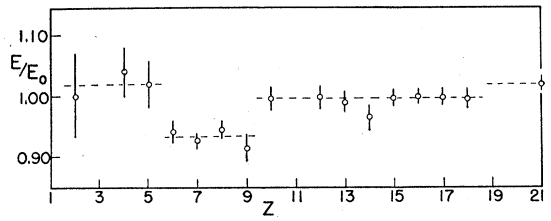

FIG. 9. Ratio of the Coulomb energy calculated by the central core model to the experimental values. $E=4.31(Z/R_0)f(\rho)$.

low energy tracks. However, the high energy end has a shape predicted by the Fermi theory.

CALCULATION OF COULOMB ENERGY

The experimental values of the Coulomb energy of the "last proton" calculated by Eq. (1) are shown in Fig. 7 as a function of atomic number. From this it is seen that the Coulomb energies of this series can be represented by a smooth function of the atomic number plus small local variations which seem to be quite random in their occurrence. However, in some cases, these variations are several times larger than the probable error of the measurements.

It has been pointed out[1,2] that the smooth curve is easily explained from the assumption that the nucleus is a homogeneous sphere the volume of which is proportional to the number of particles. This assumption leads to the following expression for the Coulomb energy of the last proton:

$$E_c=0.6(A-1)e^2/r_0A^{\frac{1}{3}},$$

where A is the mass number of the nucleus, $r_0A^{\frac{1}{3}}$ the volume of the nucleus, and e the electronic charge. If r_0 is determined from the Coulomb energy of the "extra proton" of P^{29} (5.41 Mev), this gives

$$E_c=0.592(A-1)/A^{\frac{1}{3}}\ \text{Mev} \quad \text{and}$$
$$r_0=1.47\times10^{-13}\ \text{cm}.$$

Figure 8 shows the ratios of the calculated Coulomb energies to the experimental values. It is seen that the theoretical and experimental results agree for $Z\geqslant 6$, therefore our assumption that the volume of the nucleus is proportional to the number of particles must be satisfied. However, for $Z\leqslant 6$ the curve indicates that the volume occupied per particle increases with decreasing atomic number.

Bethe[2] has proposed a model to account for the local deviations of the Coulomb energies by taking into account the extension of the wave function of the "extra proton." For this he assumes that the nucleus consists of a homogeneous core of radius R_0 and an "extra proton," the wave function of which is assumed to be a constant inside the core and to satisfy the force-free wave equation outside the core.

In the above references several approximations were made which led to quite large errors in some cases. A direct calculation leads to the following formula for the Coulomb energy of the "extra proton,"[10]

$$E_c=3Z_0e^2f(\rho)/R_0 \qquad (2)$$

$$\rho=b/R_0 : b=\hbar/2(2M\epsilon)^{\frac{1}{2}} : R_0 = r_0[A_0+1/(1+3\rho)]^{\frac{1}{3}},$$

where Z_0 is the atomic number of the "core," A_0 the number of "core" particles, ϵ is the binding energy of the "extra neutron," and f is a slowly varying function of its argument (see Fig. 10). If b, and R_0 are measured in units of 10^{-13} cm, and the parameter r_0 determined to give the observed value of the Coulomb energy of the "extra proton" of P^{29}

$$E_c=4.31Z_0f(\rho)/R_0\ \text{Mev}. \qquad (3)$$

Table I shows the values of the various variables appearing in Eqs. (2) and (3). Here

TABLE II. *Coulomb energies.*

ATOMIC NUMBER	HOMOGENEOUS NUCLEUS MODEL	HOMOGENEOUS CORE MODEL	EXPERIMENTAL VALUES
2	0.822	0.689	0.70
3	1.39	——	0.80
4	1.86	1.70	1.81
5	2.28	1.80	1.84
6	2.66	2.57	2.73
7	3.03	2.77	2.98
8	3.37	3.30	3.50
9	3.69	3.40	3.70
10	4.0	3.95	3.98
11	4.30	4.18	——
12	4.59	4.59	4.60
13	4.86	4.72	4.77
14	5.14	5.14	5.32
15	5.41	5.38	5.41
16	5.66	5.63	5.65
17	5.91	5.89	5.91
18	6.16	6.14	6.18
21	6.88	6.92	6.73

[10] This calculation has been carried out somewhat differently from that of Bethe.[2] See Appendix for details.

$Z_0=Z-1$, $A_0=2Z-1$, and $r_0=1.32$. The values of ϵ were taken from Barkas' table of masses.[11] Table II lists the observed coulomb energies together with the values calculated by each of these models.

Figure 9 shows the ratio of the values of the Coulomb energy calculated by the central core model to the experimental values. This graph has an altogether different character than the graph shown in Fig. 8, derived from the homogeneous model of the nucleus. There is no definite uptrend for the lighter nuclei and instead of the seemingly unrelated deviations of points from the average, they fall into four definite groups as shown by the dotted lines. At first one might be inclined to think that the group character of the points in Fig. 9, as well as the relatively large separations between groups, might be due to an over-correction of the model, the roughness of which cannot be denied. However, examination of Table I shows that the values of ϵ for the elements of even atomic number $Z=6$ to $Z=14$ are all about the same, and much more than the values of ϵ of their odd neighbors. If large errors are present in the model due to over-correction, then the average of the points corresponding to even Z should lie definitely apart from the average for odd Z. Although there seems to be a slight tendency toward such an effect between the elements $Z=6$ and $Z=9$, the large separation between $Z=9$ and $Z=10$ cannot be explained on this basis. Therefore, it appears that the "cores" of these elements vary in some regular manner. The "core" affects Eq. (3) only through its radius R_0, so that Fig. 9 indicates that elements having "cores" of 10, 12, 14, and 16 particles have radii smaller than given by Eqs. (2). This means that the "cores" of these elements are more tightly bound than those having, for example, 2, 6, or 8 particles in the "core." Although this is in agreement with the low binding energies of H^2, Li^6, Be^8, too much weight must not be given to the corresponding values of the nuclear radius of these elements, because the above model cannot be expected to be reliable for so few nuclear particles. However, this objection is not present for the higher mass numbers and the grouping can be attributed to abrupt changes in the average volume occupied by a neutron or proton in the "core." The fact that one of these transitions occurs after a "core" of 16 particles and the other somewhere between 36 and 40 particles is in agreement with the shell structure of nuclei predicted from the Hartree model.[11,12]

The authors are indebted to Professor Bethe for first suggesting this research and to Professor Weisskopf for further encouragement. We wish to thank also Dr. R. G. Sachs for discussions and suggestions concerning some of the theoretical aspects.

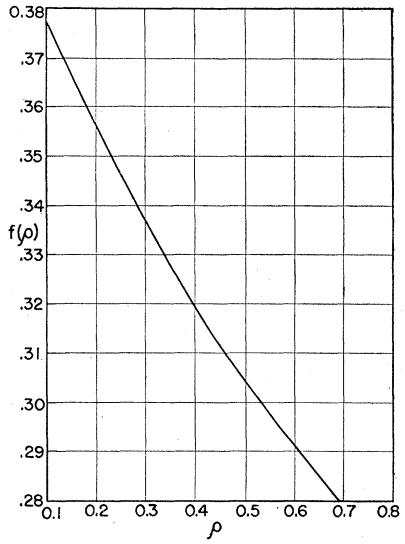

FIG. 10. Values of the function $f(\rho)=[0.4+I(\rho)]/(1+3\rho)$.

APPENDIX

According to Bethe[2] the wave function of the "extra neutron" is

$$U_r=(a/r)\exp[-(r-R_0)/2b] \quad r\geq R_0,$$
$$U_r=a/R_0 \quad r\leq R_0,$$

where $b=\hbar/2(2M\epsilon)^{\frac{1}{2}}$, and ϵ is the binding energy of the "extra neutron," to be taken from experiment, and M is the reduced mass of the system.

The normalization condition

$$\int U^2 dv=1$$

gives

$$a^2=1/4\pi(b+R_0/3).$$

If, now, the "extra neutron" is replaced by a proton, it is assumed that the wave function of the proton is the same as that of the neutron, the Coulomb energy of the

[11] W. H. Barkas, Phys. Rev. **55**, 691 (1939).

[12] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 173 (1936).

"extra proton" is given by[2]

$$E_c=\frac{Ze^2}{b+R_0/3}\left[\frac{2}{5}+\int_{R_0}^{\infty}\frac{dr}{r}\exp\left(\frac{-(r-R_0)}{b}\right)\right]. \quad (4)$$

If we let

$$\rho=b/R_0,$$

$$I(\rho)=\int_{-R_0}^{\infty}\frac{dr}{r}\exp\left(\frac{-(r-R_0)}{b}\right),$$

and

$$f(\rho)=\frac{0.4+I(\rho)}{1+3\rho},$$

Eq. (4) becomes

$$E_c=3Z_0e^2f(\rho)/R_0.$$

If b is measured in units of 10^{-13} cm then

$$b=\frac{2.36}{\epsilon^{\frac{1}{3}}}\left(\frac{A_0+1}{A_0}\right)^{\frac{1}{3}},$$

where ϵ is expressed in m.m.u., and $f(\rho)$ is shown plotted in Fig. 10.

It is now assumed that the volume of the core is proportional to the number of core particles A_0 plus the fraction of the "extra neutron" within the sphere of radius R_0. Therefore,

$$R_0^3=r_0^3\left(A_0+\int_0^{R_0}U^2dv\right)$$

or

$$R_0=r_0[A_0+1/(1+3\rho)]^{\frac{1}{3}}.$$

OCTOBER 1, 1941 PHYSICAL REVIEW VOLUME 60

A Rotational Analysis of Some CS_2 Bands in the Near Ultraviolet System*

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(Received July 7, 1941)

Six bands in the λ3100 system of CS_2, photographed in absorption on a 30-foot grating spectrograph, have been analyzed and B values are given for their initial and final states. The band structure is of the simple PR branch type, all of the observed bands originating from $^1\Sigma^+_u-{}^1\Sigma^+_g$ transitions. The bands at λ3468, λ3501, and λ3535 originate in the normal state, giving a value $r_0=1.548$A for the normal carbon-sulphur separation. A partial vibrational analysis shows that for the bending vibration, $2\nu_2''=802$ cm^{-1}. Two progressions, $v_2''=0$ and $v_2''=2$, indicate that $\nu_2'=270$ cm^{-1}. Two bands, λ3501 and λ3601, have their common upper $^1\Sigma^+_u$ state perturbed by a $^1\Pi_u$ state. A perturbation analysis gives constants which are in good agreement with observation and in addition gives the B value for the perturbing $^1\Pi_u$ state. Evidence is presented which indicates that the excited electronic state of CS_2 is bent. It is shown that, even if the molecule is bent to 125°, the P and R series may still be represented by a simple quadratic formula exactly as for a linear molecule, for J values less than 25. The state nomenclature used ($^1\Sigma^+_u$, $^1\Pi_u$, etc.) is that corresponding to the vibronic (electronic-vibrational) states of a linear molecule.

Introduction

THE analysis of the carbon disulphide spectrum has been of considerable interest to the band spectroscopist. Measurements on the Raman spectrum[1] of the liquid and on the infrared spectrum of the gas[2–4] have determined the fundamental vibration frequencies in the normal state. Recent improvements in the resolving power of infra-red spectrometers have made possible a rotational analysis of the $\nu_1+\nu_3$ band. The analysis showed it to be of the parallel type with $B''=0.112$ and $B'=0.111$.[5]

An investigation of vibrational structures in the far ultraviolet absorption spectrum has been made by Price and Simpson.[6] They were unable to make a definite assignment of vibrational frequencies of the molecule in the excited states, but reported possible series.

The first reported attempt to analyze the well-known ultraviolet absorption system near

* Assistance in the preparation of the materials was furnished by the personnel of Works Project Administration Official Project No. 665-54-3-87.

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