The Radiations Emitted from Artificially Produced Radioactive Substances

I. The Upper Limits and Shapes of the β-Ray Spectra from Several Elements*

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The β-ray spectra of N^{13}, F^{17}, Na^{24}, Si^{31}, P^{32}, Cl, A^{41} and K^{42} have been investigated by measuring the curvatures of the tracks due to the β-rays in a cloud chamber traversed by a known magnetic field. By allowing the tracks to be formed in hydrogen the scattering of the tracks is so reduced that the distribution curves are felt completely to represent the true distributions for momenta greater than $1000H\rho$. It is found that the shapes of these curves are in very good accord with the Konopinski-Uhlenbeck modification of the Fermi theory for the first five elements mentioned above (two positron emitters and three electron emitters). The spectra of the last three elements above can be resolved into two components, each of which has a shape which fits the theory. In the case of N^{13} an upper limit has been found from fitting the data with a theoretical curve at 1.45 MV. The K–U theory always indicates a higher upper limit than is found by inspection of the data, because of the high order of contact with the momentum axis which it demands. The upper limit of N^{13} can be calculated from known reaction energies and should be 1.5 MV. The excellent agreement of these two values of the upper limits is regarded as suggesting that the high K–U limits represent the true energy changes in a β-disintegration.

SINCE the discovery by Curie and Joliot[1] that several elements could be made radioactive by bombardment with α-rays others[2,3,4] have found that the same could be accomplished by the use of protons, deuterons or neutrons as the primary agents. These artificial elements are isotopes of known stable elements and are in general β-ray emitters. (We shall use the term "β-ray" to embrace both positrons and electrons.) The opportunity to study the very perplexing problem of β-emission is somewhat more favorable for these light elements, and for the following reasons: There are no subsequent products which may confuse the data; the opportunity is available to obtain independent values of the energy changes in β-emission by means of other nuclear reactions; the β-rays arise from light nuclei, which are presumably simpler; finally, it is very easy to prepare a great number of these bodies of such activities that one can do more careful work with them than is feasible with any but a few of the naturally radioactive elements.

The present work began simply as an attempt to arrive at values for the energy changes in β-emission, by measuring the radii of curvature of the tracks due to the β-rays from various deuteron-activated bodies in a cloud chamber traversed by a known magnetic field. It is generally agreed that the "upper limit" of a continuous β-ray distribution corresponds to the energy change of the reaction. This upper limit is usually practically defined as corresponding to the energy of the fastest track observed. Rigorously it must be an energy such that no track will ever be found with an energy exceeding it no matter how many tracks are measured. Naturally such a limit can be found experimentally only by extrapolation. If the distribution curve approaches the axis of momenta very steeply then it will be possible to set a value to the upper limit, by observing the fastest tracks, which may be very close to the true limit. However, if the distribution curve approaches the axis very slowly then such a practice will lead to low values of the limit. It seems to be more satisfactory to fit a theoretical curve to the data (in the form of a distribution histogram) and take the upper limit from it by extrapolation. Until recently it has been impossible to attempt this because of the absence of a suitable theory,

* Presented at the Berkeley meeting of the American Physical Society. The data given in this paper have all been recalculated and supersede anything given in the abstract of that meeting.

[1] I. Curie and F. Joliot, Comptes rendus **198**, 254 (1934); Nature **133**, 201 (1934).

[2] J. D. Cockcroft, C. W. Gilbert and E. T. S. Walton, Nature **133**, 328 (1934).

[3] M. C. Henderson, M. S. Livingston and E. O. Lawrence, Phys. Rev. **45**, 429 (1935).

[4] E. Fermi, Ricerca scientifica (1) **5**, 283 (1934).

but now a theory due to Fermi[5] together with a modification due to Konopinski and Uhlenbeck[6] is available. In getting an upper limit in this way, by fitting a theoretical curve to the data and extrapolating, it is important that the shape of the curve is not in any way falsified by experimental conditions.

Last spring we examined the Fermi theory to see if it predicted the shape of the distributions we were getting and reported favorably on the agreement between the two at the Washington meeting of the American Physical Society in April, with the reservation that we did not feel that our experiments were good enough satisfactorily to test a theory. At that time we were using oxygen as the gas in the chamber as is usual in β-ray work. In measuring the curvatures of the tracks we had adopted the rule that all tracks with visible deflections in them were to be rejected. That this was distorting the shape of the distribution curves we knew because we were being forced to discard many more of the low energy tracks than the high energy ones. This distortion can be reduced to a very great extent by photographing the β-tracks in hydrogen instead of oxygen. The scattering is thus reduced by a factor of 64 so that what little scattering remains is probably due to the heavier nuclei in the vapor.

We found with the hydrogen filled chamber that the distribution curves were more skew than they had appeared with the oxygen filled chamber. This is not surprising: our criterion of selection had been forcing us to discard as unmeasurable a large number of low energy tracks. The number discarded increased as the energy of the track decreased. The apparent concordance between our early data and the Fermi theory was entirely traceable to this because the Fermi distribution function is very nearly symmetrical, so that when the number of low energy tracks measured was increased this apparent symmetry in the experimental distributions was lost. At the suggestion of Dr. Arnold Nordsieck we attempted to fit our new data to the then unpublished modification of the Fermi theory due to Konopinski and Uhlenbeck. The K–U theory has the here desirable feature of asymmetry and was found to fit the experimental data very well. This symmetry carries with it, however, a high order of contact of the distribution curve with the momentum axis, which leads to an upper limit greater than the last measured track in most cases. Because of this we hesitated to attribute any physical significance to these higher upper limits. Recently, however, the reaction energy pertaining to a β-ray change has been established in one case (N^{13}, see later) independently of any β-ray measurements. This energy change is in excellent agreement with that indicated by fitting a K–U curve to the data on the β-ray spectrum.

We have reported on the excellent way in which the K–U theory accounts for the shape of the curves in a Letter to the Editor of this journal[7] and also in the nuclear symposium at the Los Angeles meeting of the American Physical Society, without urging the acceptance of the higher upper limits indicated by it.

In this paper we shall present distribution curves of the β-rays emitted from deuteron-activated carbon, oxygen, sodium, silicon, phosphorus, chlorine, argon and potassium. With them the K–U theory will be tested insofar as the shape of a distribution curve is concerned. From fitted theoretical curves we shall deduce values for the various endpoints, which we feel little hesitancy in offering as representing the energy change in the β-emission.

Experimental Details

With the magnetic resonance accelerator of Lawrence and Livingston[8] it has been possible to stimulate radioactivity in elements as heavy as platinum[9] using an intense deuteron beam of as much as 15 microamperes at as high as 5.3 MV. Depending upon the nature of the radioactive substance it is desired to create, a suitable target is bombarded in a vacuum or in air (the deuteron beam emerges through a thin window into a "target chamber") for a time sufficient to give

[5] E. Fermi, Zeits. f. Physik **88**, 161 (1934).
[6] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **48**, 7 (1935).
[7] F. N. D. Kurie, J. R. Richardson and H. C. Paxton, Phys. Rev. **48**, 167 (1935).
[8] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 608 (1935).
[9] J. M. Cork and E. O. Lawrence, Am. Phys. Soc., Berkeley Meeting, Phys. Rev. **49**, 205 (1936).

a convenient activity. In many cases it is desirable to protect the target from contamination in the target chamber by covering it with a thin foil, usually platinum. A great variety of targets have been used. The usual one consists of a fused salt of the element to be studied, the other component of the salt being generally one with either a very short half-life or with a low activity. Thus for studying radio-sodium it is convenient to use NaF, since the half-life of the fluorine activity (10 sec.) is much shorter than that of sodium (15 hours). Such a target is always "aged" for a sufficient time to allow the activity in the other component to decay to an insignificant magnitude. Similarly radio-chlorine may be examined in the form of $PbCl_2$ since the heavy element lead is activated to a much less degree if at all. Where possible the pure element is used as a target. Even gases may be bombarded by flooding the target chamber with them and collecting the radioactive elements by recoil on a piece of platinum foil.

The activated target is then put in one or the other of two holders mounted near the periphery of a Wilson chamber. The first and more convenient of these holders consists of a brass well set into the roof of the chamber with a grid, also of brass, covered by a thin mica window cemented on with gelatin. We have used this container for most of our work, because the cloud chamber can be kept clean and uncontaminated and sources can be dropped into it and photography begun immediately if desired. We have felt that there were objections to this holder for the following reasons: The mica window is so large that no definite solid angle for the β-rays is defined, the solid angle depending on the radius of curvature of the track and hence on its energy. (From a comparison of the shape of the distribution curves obtained with this holder and with the other (see later) which limits the solid angle more uniformly for all energies the conclusion has been reached that this effect is not serious.) The mica window had a thickness of about 8 mg/cm^2 and so caused a retardation of at most 60–80$H\rho$. The brass grid itself is only about 800 mg/cm^2 thick and so will allow electrons of 2 MV and over to penetrate it. This causes no trouble except for high energy spectra, such as the spectrum of chlorine, and here we do not rely on the data obtained with this holder alone.

The second holder was designed to eliminate these objections. It consists of a half cylinder of aluminum with an equatorial slot which confines the β-rays to the central plane of the cloud chamber. The walls of the aluminum slit are over 1500 mg/cm^2 thick and so will stop all β-rays of energy below 3 MV. It is made in the form of a half cylinder so that there will be no heavy backing to cause undue reflection of the electrons. This holder is something of a nuisance to use since the cloud chamber has to be taken down to insert a source and considerable time elapses before photography can begin. In general therefore we use the first holder for we have found that its shortcomings are not important: their influence on the shape of the distribution curve is only felt at the very low energies where all data are doubtful; the upper limit obtained is apparently independent of the holder. We shall mention as we consider the separate elements with what holder the data have been taken.

The cloud chamber is the same one mentioned by one of the authors previously.[10] It is 2 cm deep and 16 cm in diameter. For all the work presented in this paper it was filled with tank hydrogen and operated at about 100 cm pressure. The vapor was isopropyl alcohol. The tracks were illuminated by a shorted carbon arc and photographed with a single Sept camera equipped with an $f2$ Schneider lens of 50 mm focal length.

Above and below the cloud chamber are two coils of the following approximate dimensions: I.D. 8.36 inches, O.D. 9.93 inches, height of each coil 2.5 inches, separation of the two coils 2.56 inches. The coils are wound on a brass form, each carrying 829 turns of No. 18 wire. The two coils are connected in parallel and have a constant of 33.5 gauss per ampere. This figure has been obtained by comparing the deflections of a ballistic galvanometer when a standard search coil whose area-turns are known to less than 0.2 percent "by construction" was placed in the field of the chamber coils and when it was placed in the field of a standard solenoid whose constants were also known to less than 0.1 percent "by construction." All the values have been

[10] F. N. D. Kurie, Phys. Rev. **47**, 97 (1935).

checked against a standard mutual inductance of 50 millihenrys and agree to 0.3 percent. We regard our field as being accurately known to one percent at the center of the chamber. The ammeter by which the current is read during the photographing of a series of expansions has been calibrated from time to time against a standard cell and standard 0.1-ohm resistance and has been found to maintain its calibration within the accuracy to which it can be read (about one percent). The coils are closer together than the Helmholz position but over the region where tracks are formed the field is uniform to less than four percent. This would mean a maximum distortion of the measured curvature of a track of two percent if the field fell off linearly. Actually it falls off much less rapidly until the periphery of the chamber. Hence this effect can account for an error of approximately one percent in the radii of the tracks.

The time which the coils require to reach their maximum current has been estimated with a cathode-ray oscilloscope at between 1/120 and 1/60 sec. Since the coils are turned on more than a second before the chamber expands the field has ample time to reach maximum strength before the chamber is photographed.

On 110 volts d.c. the coils produce a field of about 500 gauss. They may also be used up to 220 volts giving a field of 1000 gauss. This range of fields covers β-rays from $1000H\rho$ to around $25{,}000H\rho$ (0.1–7 MV) with curvatures of an easily measurable magnitude. In order to prevent the coils from overheating the current is passed through them for only about two seconds during the expansion of the cloud chamber. At the high field the heating of the coils is sufficient to require the chamber to be water cooled in order that tracks be undisturbed. In general, however, the 500 gauss field is sufficient to cope with the β- and γ-rays encountered, at which field there is no thermal disturbance at all.

The film after development is replaced in the camera and reprojected on a screen placed at the same optical distance from the camera as the cloud chamber was when the pictures were originally taken. The tracks are measured by comparing them with a set of circles engraved on a sheet of celluloid. The circles have radii from 1 to 29 cm drawn at intervals of 0.5 cm. The error in measuring a curvature is less than five percent in the region of the upper limits. However, since it is our practice to plot the data in class intervals of either 500 or $1000H\rho$ this error does not appreciably alter the shape of the distribution curves. The use of hydrogen in the chamber so diminishes the scattering of the tracks that except for the very slowest (less than $1000H\rho$), which are in general not measured, few deflections are noticed in the tracks. Indeed it is an exceptional track which has a discernible deflection. The tracks in hydrogen are resolved into individual droplets; this makes them easier to measure than the broader tracks in oxygen, for one can fit the circles on the celluloid scale to the reprojected track by trying them for a bisection of the images of the droplets. This seems to us to be a more precise method of circle fitting than can be achieved with the oxygen tracks. Despite the very large reduction in scattering experienced with hydrogen occasional tracks which have had their curvatures falsified by small deflections will find their way into the final data, and this must be borne in mind in evaluating such data. Probably this effect is no worse than the everpresent chance in a magnetic spectrograph that a particle which has had many reflections from slits will be counted when a portion of the distribution curve which contains few particles is being examined.

The tracks are plotted as a histogram showing the number of tracks in a given $H\rho$ interval or in a given radius interval (either 0.5 cm or 1 cm depending on the number of tracks available). The latter is done in some cases rather than to plot against $H\rho$ directly to obviate systematic irregularities which may be introduced into the curve. Thus at 540 gauss the interval 1300–$1400H\rho$ will include only tracks of 25 cm radius while 1200–$1300H\rho$ will take in tracks of 23 and 24 cm radius and likewise 1400–$1500H\rho$ will contain 26 and 27 cm radius tracks. There will consequently always be a deficiency of tracks in the interval 1300–$1400H\rho$. For some field values this artificial irregularity does not exist, but when it does we have adopted the practice mentioned above and merely mark on the graph the appropriate $H\rho$ values. The relation between the $H\rho$ of a given track and its energy in MV is given by the following formula, which is valid

for all values of the energy:

$$H\rho=(10^4/3)(E(E+1.02))^{\frac{1}{2}}.$$

The number 1.02 is simply twice the rest mass of an electron expressed in millions of volts.

The histogram giving the frequency distribution in $H\rho$ may be transferred into a distribution in energy by dividing point by point each ordinate by the velocity of the electron.

Because of the difficulty of measuring small radii of curvature together with the increased number of rejections due to the augmented frequency of deflections in the tracks of low energy β-particles the portion of the histogram referring to them is in general stunted. With hydrogen and careful measuring this extends up to about $1000H\rho$, with oxygen it is apparent up to $4000H\rho$ and beyond. If the body being studied emits γ-rays along with its electronic radiations secondary electrons will be mixed with the disintegration β-particles, and will, if the γ-ray energy is greater than the β-ray upper limit, add a tail to the distribution which will again lead to a false value of the upper limit. These difficulties must be kept in mind in dealing with cloud chamber data.

We shall see that on the basis of any data on β-ray emission which we now possess the Konopinski-Uhlenbeck modification of the Fermi theory completely describes the process of the emission of a β-ray so that we may assign upper limits to the spectra by fitting to them a K–U curve.

Application of Theory to the Experimental Data

E. Fermi has given a theory of β-decay in which the emission of an electron by a nucleus is imagined as being occasioned by the transition of a nuclear neutron into a proton with the birth of an electron-neutrino pair. These two then escape from the nucleus with a constant total energy and the energy distribution of the electrons merely expresses the manner in which this total energy is shared between electron and neutrino. Theoretically this distribution depends upon the choice of interaction energy between the electron-neutrino field and the heavy particle. This interaction "Ansatz" is rather arbitrary and Konopinski and Uhlenbeck have shown[11] that several forms of the distribution function can be written down. They denote them by referring to the order of the derivatives of the electron and neutrino wave functions which enter into the coupling energy. Thus the (0,0) distribution is Fermi's original form; (0,1) is the form proposed by Konopinski and Uhlenbeck in their first paper. The general shape of the curves which we have experimentally found leads us to reject all the other forms which these authors have proposed, indeed, we shall see that only the (0,1) form requires consideration.

Eq. (40) of Fermi's paper gives the probability that an electron of momentum $mc\eta(\eta=H\rho/1700)$ in $d\eta$ be emitted. On integrating this from $\eta=0$ to $\eta=\eta_0$ one gets an expression for the half-life τ, if η_0 refers to the upper limit of the momentum distribution curve. Dividing the first of these two equations by the second and multiplying by the total number of events being considered (=number of tracks measured, in this case) one gets for the number N of particles with momenta between $mc\eta$ and $mc(\eta+\Delta\eta)$

$$N\Delta\eta=Kf(Z,\eta)(C-(1+\eta^2)^{\frac{1}{2}})^2\Delta\eta, \qquad (1)$$

where K is a function of Z, τ and η_0; $C=(1+\eta_0^2)^{\frac{1}{2}}=E_0+1$. In this latter expression E_0 is the energy in mc^2 units of the upper limit; this we shall use instead of $(1+\eta_0^2)^{\frac{1}{2}}$ for typographical simplicity. Fermi has given the form of $f(Z,\eta)$ explicitly and it may easily be shown that it can be reduced to the expression

$$f(Z,\eta)=\eta^2\left(\frac{2\pi y}{1-e^{-2\pi y}}\right) \qquad (2)$$

for all light elements up to about copper ($Z=29$). In (2) $y=Z(1+\eta^2)^{\frac{1}{2}}/137\eta$. We may now write (1):

$$K(N/f)^{\frac{1}{3}}=C-(E+1) \quad \text{(Fermi)}. \qquad (3)$$

It is clear that if one plots $(N/f)^{\frac{1}{3}}$ against $(E+1)$ one should get a straight line if the Fermi theory is being obeyed. Furthermore the upper limit E_0 is given by the intercept C on the $(E+1)$ axis.

The modification of the Fermi theory introducted by Konopinski and Uhlenbeck merely changes the squared term in (1) into a fourth

[11] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **48**, 107 (1935).

power so that we may write for the equation of the distribution curve:

$$K(N/f)^{\frac{1}{4}}=C-(E+1) \qquad \text{(K–U)}. \qquad (4)$$

Treating the data in an exactly analogous way will yield through C the K–U upper limit.

Results

Carbon N^{13} (positron emitter)

We have studied the radioactivity induced in carbon by fast deuterons in both the forms of Acheson graphite and soot deposited on a thin piece of copper. The first source holder was used here. To get a sufficiently strong sample requires an exposure of only a few seconds to the deuteron beam (2 microamperes at 4 MV). To avoid contamination with O^{15} and F^{17} which would be formed in the surrounding air and driven by recoil onto the carbon target the target chamber was evacuated during bombardment and further the carbon was protected by a thin copper foil (0.0001″ thick). These precautions were found to be insufficient, for a freshly activated sample gave an upper limit which was around 200 kv higher than that got from a target which had been allowed to decay through eight half-lives of N^{13} (eighty minutes). Dr. S. N. Van Voorhis in some unpublished work has found a short period of between 2 and 3 minutes in carbon activated under these conditions. This is certainly the "air effect" due to occluded air in the carbon. Indeed the high energy tracks are all to be found in the early pictures taken of a freshly activated sample. The high energy tracks have been attributed to O^{15} (and perhaps to a much smaller degree to F^{17}) on the basis of some unpublished measurements on the positron spectrum of this element by Delsasso, Fowler and Lauritsen which they have kindly communicated to us. They find that the upper limit of O^{15} is slightly greater by an amount which is about the same as we find freshly activated carbon to exceed N^{13}.

The data on old samples of activated carbon are shown in Fig. 1 and refer to 723 tracks. Through the histogram has been drawn a smooth curve which is taken from the K–U theory by fitting a straight line to the linear plot as was discussed in the previous section. This linear plot is given in Fig. 2. On it one will notice that the intercept which we have denoted by C is not beyond the last measured track. This is not usual, as we shall see, but may easily be attrib-

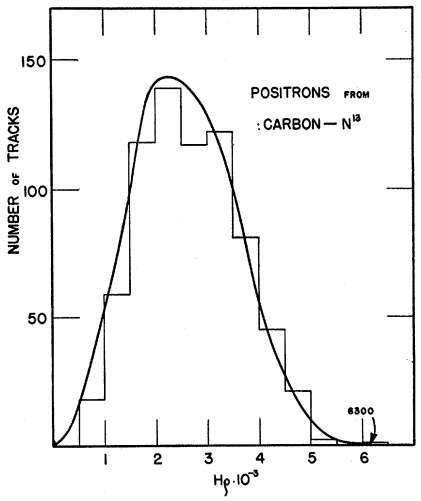

FIG. 1. Distribution histogram for the positrons emitted from activated carbon (N^{13}). Superposed on the experimental histogram is the K–U curve which has been fitted to it. The endpoint of this fitted curve is at $6300H\rho$ as indicated.

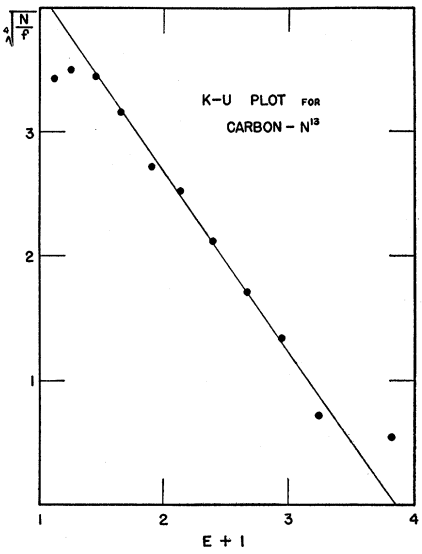

FIG. 2. K–U plot for activated carbon. Plotted in this way the points should fall on a straight line if the distribution curve has the shape demanded by theory. Deviations from linearity are here seen only at the low energy and where one always counts too few tracks, and at the high energy end where statistical fluctuations are important. The upper limit is here given by $E+1=3.8$ ($=6300H\rho$ $=1.45$ MV).

uted to errors in measurement. It is obvious from the form of (4), since the fourth root there occurring requires a high order of contact of the distribution curve with the axis of abscissae, that the theoretical curve may have an end point quite beyond the last measured tracks. It is not inconceivable that the true distribution curve should have this high order contact and that the measured "upper limit" should fall short of the true upper limit because of the inherent difficulties of following the curve to its limit. The hint that the curve might go on clinging to the axis for some distance beyond the last observed track has been given some foundation in the recent work of Scott[12] who has exhibited a curve for Ra E which passes beyond the accepted limit of $5500H\rho$. We feel some hesitancy in offering this as evidence since Scott's work was done with a magnetic spectrograph where scattering renders the counts in regions where particles are scarce very uncertain.

The evidence is good that the radioactive body in this case is formed by the following reaction

$$C^{12}+H^2=N^{13}+n^1+Q_1$$

and one must imagine it decaying according to

$$N^{13}=C^{13}+e^+ +(e^-)+Q_2.$$

A further reaction is known, namely,

$$C^{12}+H^2=C^{13}+H^1+Q_3.$$

From this set of reactions we may eliminate the heavy particles and write

$$Q_2=(H^1-n^1)+(Q_3-Q_1)-(e^+ +e^-).$$

The difference (H^1-n^1) is best taken from Feather's work[13] on the photoelectric disintegration of the deuteron and amounts to -0.48 MV. Bonner, Delsasso, Fowler and Lauritsen[14] have measured all the Q's under almost identical conditions and find $Q_1=-0.37$ MV, $Q_3=2.65$ MV and $Q_2=1.45$ MV. Taking these figures together with their probable errors we find that $Q_2=1.52\pm0.11$. From the intercept C of Fig. 2 we get $Q_2=1.45$ MV which is in excellent agreement with the result of Bonner, Delsasso, Fowler and Lauritsen and with the calculated endpoint. The agreement would not have been nearly as good if we had taken the upper limit at $5500H\rho(=1.25$ MV) which is at the point the histogram (Fig. 1) seems to indicate. It seems then very likely that the K–U theory offers a good description of the shape of β-ray distribution curves both as concerns the shape of the curve and as regards the endpoint.

Cockcroft, Gilbert and Walton[15] have published a distribution curve suggesting the existence of two groups of positrons, when carbon is activated with protons. It will be interesting to investigate this matter further inasmuch as the deuteron activation gives clearly a simple spectrum.

We have also examined the spectrum of carbon activated at energies below 1 MV. It might be expected that if the apparent simplicity of the N^{13} spectrum were due to the superposition of many elementary distributions due to different excited states of N^{13} one might alter this appreciably by the fourfold change in bombarding energy. No change was apparent in the distribution curve or the upper limit. This examination could only be made on a fresh sample because of the weak activity at the lower bombarding voltage, so the distribution curve was compared with that of another fresh sample activated at the high voltage. The upper limits obtained in these cases pertain largely to O^{15} and we can definitely say that any effect dependent on bombarding voltage is small for these elements.

Oxygen F^{17} (positron emitter)

H. W. Newson has recently shown[16] that a radioactive substance can be prepared by bombarding oxygen with deuterons and that this substance is probably F^{17}, the same element which has been prepared by Wertenstein from nitrogen with α-particles. Newson gives the half-life as 1.16 minutes. In our first source holder we have studied this activity which is also a positron emission, with a target prepared by placing a platinum foil in the deuteron beam

[12] F. A. Scott, Phys. Rev. **48**, 391 (1935).
[13] Report of the British Association discussion on nuclear physics at Norwich, Nature **136**, 467 (1935).
[14] T. W. Bonner, L. A. Delsasso, W. A. Fowler and C. C. Lauritsen, Berkeley Meeting, Am. Phys. Soc., Phys. Rev. **49**, 203 (1936).
[15] J. D. Cockcroft, C. W. Gilbert and E. T. S. Walton, Proc. Roy. Soc. **A148**, 225 (1935).
[16] H. W. Newson, Phys. Rev. **48**, 790 (1935).

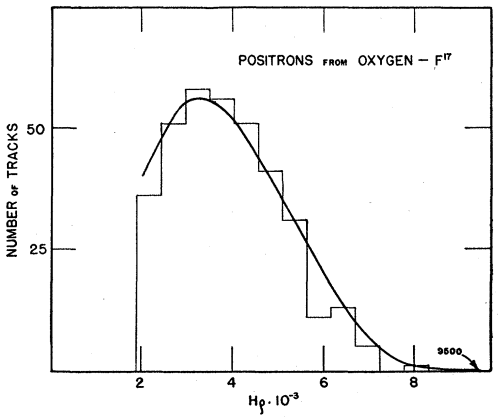

FIG. 3. Distribution histogram for the positrons emitted from activated oxygen (F^{17}) together with the theoretical K–U curve. The endpoint of the theoretical curve is at $9500H\rho$.

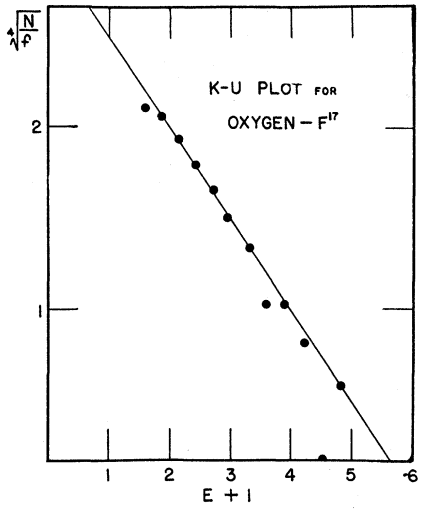

FIG. 4. K–U plot for activated oxygen. This plot is again linear and extrapolates to an upper limit at $E+1=5.7$.

when the target chamber was full of oxygen. A strong sample of F^{17} is driven by recoil onto the foil.

Fig. 3 shows the distribution histogram together with a K–U curve which has been fitted to it by means of the linear plot in Fig. 4. The upper limit indicated by the latter is 2.4 MV, while an inspection of the histogram would lead to about 2.1 MV. In this case we have again a triad of reactions such as was found in the case of carbon. These are:

$$O^{16}+H^2=F^{17}+n^1+Q_1,$$
$$F^{17}=O^{17}+e^+ +(e^-)+Q_2,$$
$$O^{16}+H^2=O^{17}+H^1+Q_3.$$

Newson estimates Q_1 as -1.8 MV. Cockcroft and Walton[17] have observed a group of protons with range 8 ± 1 cm when oxidized tungsten is bombarded with deuterons, which group vanishes when the tungsten is heated. If this group is to be attributed to oxygen according to the last of our three reactions we find $Q_3=1.8$ MV. As before $Q_2=-0.48+1.8+1.8-1.02=2.1\pm0.24$. This figure is in better accord with the value obtained from an inspection of the histogram than with that indicated by the K–U plot. The precision of the work on which Q_1 and Q_3 rest is not sufficiently great for this to be a matter of serious concern as yet.

[17] J. D. Cockcroft, Int. Conf. Physics, p. 124.

Sodium Na^{24} (electron emitter)

Radio-sodium has been extensively studied by E. O. Lawrence.[18] Substances of the same decay period have been prepared by Fermi and his collaborators[19] from magnesium and aluminum. M. C. Henderson[20] has reported the creation of the same substance from magnesium by deuteron bombardment.

We have examined the radiations emitted from this body by bombarding a target of sodium fluoride with deuterons. The activity due to the fluorine has a very short life of the order of ten seconds. The sodium targets are always "aged" before being put in the chamber. The data we have obtained from one target, bombarded for 1 hour at 1 microampere at 3 MV and aged for 4 hours were illustrated in an earlier communication from us on this subject[21] and reference is made to the figure there reproduced. That figure was based on 900 tracks, photographed with the source in our first holder. Since the time of that note we have carefully recalibrated our magnetic field as explained above and the figures for $H\rho$ of the upper limit there given must be lowered by four percent. This gives an upper limit (K–U) at $8000H\rho$ or 1.95 MV. By in-

[18] E. O. Lawrence, Phys. Rev. **47**, 17 (1935).
[19] E. Fermi, E. Amaldi, O. d'Agostino, F. Rasetti and E. Segrè, Proc. Roy. Soc. **A146**, 483 (1934).
[20] M. C. Henderson, Phys. Rev. **48**, 855 (1935).
[21] See footnote 7.

spection one would put this figure lower at, say, 1.7 MV.

In sodium we get tracks beyond the K–U upper limit but in view of the strong γ-radiation emitted from this body we attribute these tracks to the high energy component of this γ-radiation.[22]

There is some suggestion in the form of the histogram for sodium that there might be a low energy group of β-particles. Whether or not such exists is a subject for a future investigation.

Silicon S^{31} (electron emitter)

The production of S^{31} from silicon has been achieved by Fermi with neutrons and by Newson[23] with deuterons. The two authors give 145 min. and 170 min., respectively, for the half-life. The data given by us[22] earlier were obtained from a piece of quartz which had been bombarded with deuterons for 45 min. at 0.8 microamperes at about 3 MV. The sample was aged for 15 minutes. The momentum distribution curves are shown in the note referred to above. Again the magnetic field must be corrected to take account of our recalibration thus giving an upper limit at $8400H\rho$ or 2.05 MV, whereas the histogram apparently ends at about $7600H\rho$ (1.8 MV).

Phosphorus P^{32} (electron emitter)

Fermi first demonstrated that P^{32} could be made from sulphur and chlorine by neutron bombardment. Newson[23] then showed that the same substance could be made from phosphorus with deuterons, and recently Fahlenbrach[24] has produced it from the action of α-particles on silicon. The last two authors give 14.5 days and 14.0 days, respectively, for its half-life. Ambrosen[25] has made measurements on the energy distribution of the bodies produced from neutron bombardment and has shown them to be identical within the precision of his measurements. He gives 2 MV as the upper limit.

We have investigated the β-spectrum of P^{32} very carefully because it has seemed to be the ideal substance whereby to test the Konopinski-Uhlenbeck theory. It has a life which is so much longer than any other artificially produced radioactive body that contamination effects can be given ample opportunity to die out. Furthermore the only γ-radiation emitted is a soft Bremsstrahlung.

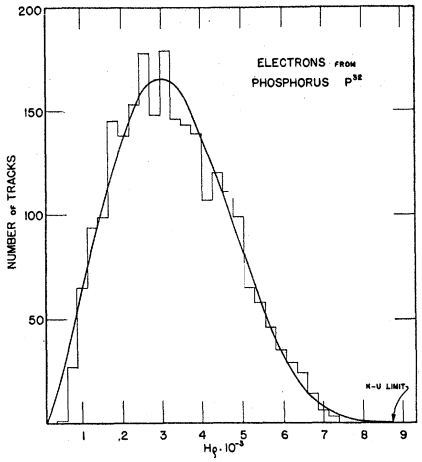

FIG. 5. Distribution histogram for the electrons emitted from activated phosphorus (P^{32}) with the fitted K–U curve showing an endpoint at $8700H\rho$.

Red phosphorus was activated in a current of 4.5 microamperes slightly under 4 MV for 2.5 hours. The sample was not used for these measurements until three days later, and then was found to be much too strong so a fragment of the 0.0001″ copper foil which had been used to protect the target chamber of the big accelerator from spattered phosphorus was found to carry sufficient activity for the measurements. This piece of foil was placed in the second holder—the experimental conditions being then as near ideal for an analysis of the shape of a distribution curve as possible. In the first place the target was sufficiently old to exclude contamination to a great extent; secondly, the β-rays were collimated so that the solid angle available was the same for all energies; thirdly, the target was thin and there was no backing except the wall of the chamber which was at least 2 cm distant so that the chance of reflection was very slight. Despite these good conditions the distribution curve differed very slightly from those we had been getting with the first source holder. On the basis of this we feel that at least for low energy spectra the first holder with the mica window on the brass grid is

[22] J. R. Richardson, Berkeley Meeting, Am. Phys. Soc., Phys. Rev. **49**, 203 (1936).
[23] H. W. Newson, Phys. Rev. **48**, 482 (1935).
[24] H. Fahlenbrach, Zeits. f. Physik **96**, 503 (1935).
[25] J. Ambrosen, Zeits. f. Physik **91**, 43 (1935).

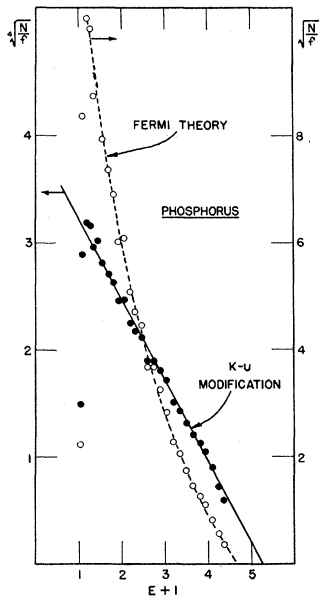

FIG. 6. The data given by the histogram in Fig. 5 are here plotted in two ways: The (black) points marked "K–U modification" should fall as they do on a straight line if the K–U theory is being followed. If the Fermi theory is being followed the (white) points should fall on a straight line as they clearly do not. This curved line has been found for all radioactive bodies when plotted in this way and indicates the character of the observed deviations from the simple Fermi theory. The K–U line shows an endpoint at $E+1=5.2$.

sufficiently satisfactory for upper limit determinations, and gives the shape of the distribution curves accurately above $2000H\rho$.

Fig. 5 shows a distribution curve containing 2372 tracks together with a theoretical K–U curve obtained from the linear plot in Fig. 6. The Fermi plot also shown in Fig. 6 is typical of the curves we began getting as soon as we substituted hydrogen for oxygen in the cloud chamber. Here the data have been plotted so that if Eq. (3) were being obeyed the line marked "Fermi theory" should be straight. The concavity shown here is to be found in all of our hydrogen-chamber distributions and is characteristic of the observed deviations from the Fermi theory. On the other hand the line marked "K–U modification" should be straight if the K–U theory is being followed. The plotted points have had a straight line drawn through them and we regard the closeness with which the points cluster around this line as a measure of the very excellent way in which this theory accounts for the shape of a β-ray distribution curve. The fact that the K–U line in Fig. 6 cuts the axis beyond the last experimental point has been a strong argument against the acceptance of the upper limits given by fitting the data in this way. However, it must be realized that the intensity in this extended tail is very small. Thus in the present case the chance of finding a track with a momentum of $8400H\rho$ can be shown on the basis of the K–U curve which we have fitted to our data to be about 1 in 4000. Also the complete agreement between the upper limit of the positron spectrum from N^{13} and the energy computed from other reaction energies makes it seem very certain that these K–U upper limits are the significant endpoints from the standpoint of energy balances.

The upper limit indicated by the K–U plot is at $8700H\rho$ ($=2.15$ MV) whereas one would estimate from the histogram at about $7500H\rho$ ($=1.8$ MV). Either figure is in good agreement with the limit of 2 MV found by Ambrosen.

Paxton[26] has reported the emission of positrons from a target of deuteron activated red phosphorus. The half-life of the active body in this case is around 50 hours and so is not associated with the electronic emission from P^{32}. A more detailed account of this work will be published shortly.

Chlorine (electron emitter)

An active chlorine was prepared by Fermi by bombarding chlorine with neutrons. What is presumably the same body has been made in this laboratory and various workers have given the half-life values ranging from 39 min. to 40.5 min. as compared with that found by Fermi of 37 min. A sample of this substance was prepared by bombarding AgCl, fused on a piece of silver, with about one microampere of 3 MV deuterons for 20 minutes. This was examined in the first source holder and a curve obtained which was very strongly asymmetric showing a strong maximum at around $2500H\rho$ and an endpoint beyond $18{,}000H\rho$. We had always got such curves from chlorine before and thought that the grid on which the mica window was supported might be slowing fast electrons thus causing them to be grouped among the slower ones. A K–U plot of

[26] H. C. Paxton, Berkeley Meeting, Am. Phys. Soc., Phys. Rev. **49**, 206 (1936).

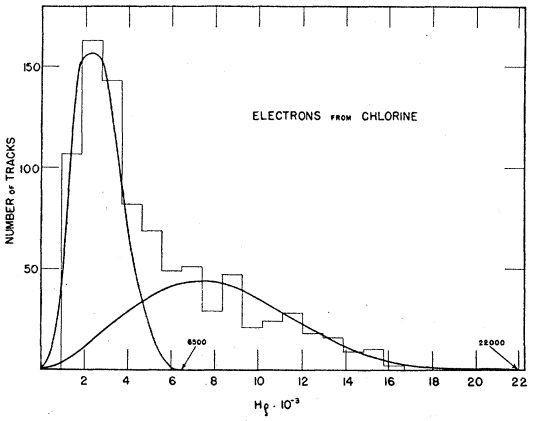

FIG. 7. Distribution histogram for the electrons emitted from activated chlorine. The general shape of this curve is much more skew than a simple K–U curve. As can be seen from Fig. 8 one may resolve this curve into the sum of two simple K–U curves. The upper limits so found are at $6500H\rho$ and $22{,}000H\rho$.

the data was found to be resolvable into two, suggesting that radio-chlorine might either consist of two separate active bodies with the same half-life or that its spectrum might be the sum of two simple P^{32}-like spectra, as Ellis and Mott have suggested for the heavy radioactive bodies.

This point is so interesting that we activated another sample and allowed it to age for three hours before mounting it directly in the chamber with no grid in front of it. The distribution curve for this case is shown in Fig. 7 and the K–U plot in Fig. 8. Here again the possibility of separating the curve of Fig. 7 into the sum of two was again apparent. When this was done upper limits of the two groups were indicated at the same points as had been obtained in the preceding case when the brass grid was in place. The relative proportions of the high and low energy groups with the grid was roughly 2 : 1 and without the grid 1 : 1. We feel that this is probably to be accounted for by the fact that the absorption curve is much steeper for electrons of the energies which make up the lower group than for the upper group.

Feeling that the legitimacy of such a resolution of the data into groups was perhaps questionable we did not pursue the matter further. It was after this that the data on phosphorus and the agreement of the reaction energies in the case of N^{13} added so much weight on the side of the K–U theory. Such a separation now seems to be a perfectly valid consequence of the apparently good accord between theory and experiment, and we plan to investigate this matter more carefully later.

If this resolution is right we may think of active chlorine decaying in one of two ways to form a stable argon. There should then be γ-rays of energy equal to the difference. The K–U plot in Fig. 8 gives the two β-ray limits as 1.5 and 6.1 MV. The difference is 4.6 MV. Richardson[22] has reported preliminary data which indicate two γ-lines at 2.4 and 1.9 MV. It might be that the transition from the excited level at 4.6 MV in argon to the ground state might be accomplished in two groups of $2.4+1.9=4.3$ MV. Unfortunately Richardson finds these lines to differ in intensity, but the data are as yet not sufficiently good to be sure the above explanation is invalid. Another alternative is to assign one group to Cl^{36} and the other to Cl^{38} attributing the fact that the periods are identical to coincidence. Again one might imagine the two groups being due to two successive disintegrations of which the second had a period short compared to the first. Against this may be raised the objection that if the active body is really a chlorine isotope then any argon isotope which could result is stable. None of these are very satisfactory and so we defer the question until more data has been accumulated.

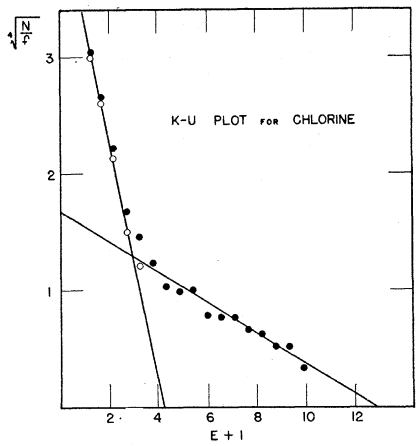

FIG. 8. K–U plot for activated chlorine. The black points are the experimental data. The presence of two linear portions to this curve is apparent. The lower group is indicated by the white points which are got by subtracting the "background" due to the lower energy part of the upper group. Endpoints are given by $E+1=4$ and 13. (The scale of abscissae in this figure has not been changed to conform to our recalibration of the magnetic field and so shows endpoints higher than the ones quoted.)

It is possible that such a presumably composite spectrum as this would exhibit a dependence on bombarding voltage. The opportunity to examine this has not arisen as yet.

Argon A^{41} (electron emitter)

That activity could be excited in argon by deuterons was discovered by A. H. Snell[27] who gives it a period of 108 minutes. Dr. Snell has prepared for us sources of this body by recoil on a platinum foil immersed in an atmosphere of argon through which the deuteron beam was passing. Photographs were taken with the foil mounted in the second source holder. The results of these photographs are shown in Fig. 9 as a distribution histogram. This has a shape very similar to the curves found for chlorine and indeed the K–U plot in Fig. 10 suggests that here again one might think of this curve as being decomposable into two. In this case it seems that the data are too few to warrant setting a value for the upper limit of the faster group, except to say that it is in the neighborhood of 18,000$H\rho$ (=5 MV) according to the K–U plot. The lower group would fall at 6600 $H\rho$ (=1.5 MV). These seem to be unrelated to the γ-ray at 1.3 MV which Richardson[22] has found, but it is a little premature to discuss this too fully until more complete data are available.

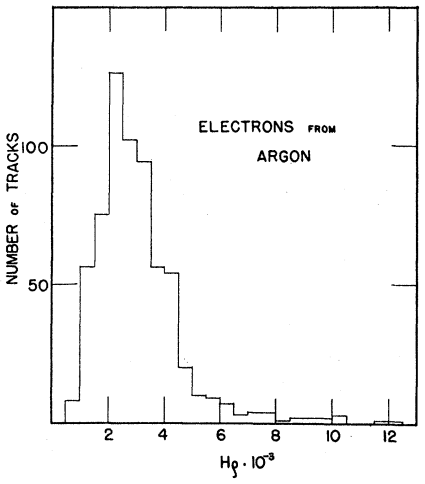

FIG. 9. Distribution histogram for the electron emitted from activated argon (A^{41}). This curve also shows evidence in its skewness of the existence of a high energy group. The data on this upper group are not abundant enough to warrant the drawing of theoretical curves on this histogram.

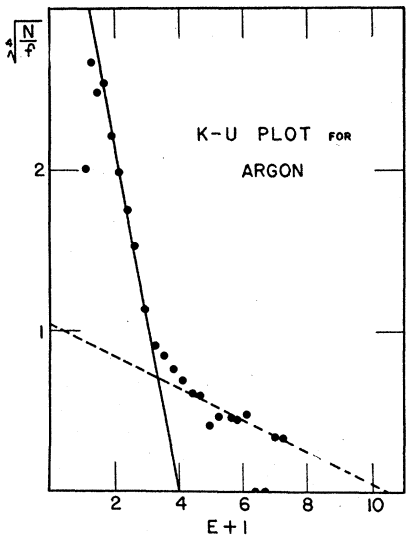

FIG. 10. K–U plot for activated argon. The lower group has an upper limit at $E+1=4$ (=1.5 MV) and the tentative trend of the upper group has been indicated by the dotted line. The endpoint of this upper group is around $E+1=10.5$ (=5 MV).

Potassium K^{42} (electron emitter)

This element was first prepared from potassium by Fermi with neutrons and later by Hevesy[28] from scandium with neutrons and in this laboratory with deuterons. This is a substance for which we also found evidence of two groups in the oxygen filled chamber. The target responsible for the 870 tracks whose momentum distribution is shown in Fig. 11 was potassium fluoride fused on a piece of platinum gauze. It was activated for one hour with one microampere of about 3 MV deuterons. In Fig. 11 we have again resolved the distribution into two groups as before. The hint to do this is contained in Fig. 12 where the K–U plot is fitted very closely by two straight lines and not at all by one. The upper limits are at 6300$H\rho$ and 16,100$H\rho$. The lower group is estimated from these data (which were taken with the first source holder, and hence are not greatly to be trusted) at half the intensity of the upper group. We have attempted to investigate the rather weak γ-radiation emitted by this element but have not

[27] A. H. Snell, Berkeley Meeting, Am. Phys. Soc., Phys. Rev. **49**, 207 (1936).

[28] G. Hevesy, Nature **135**, 96 (1935).

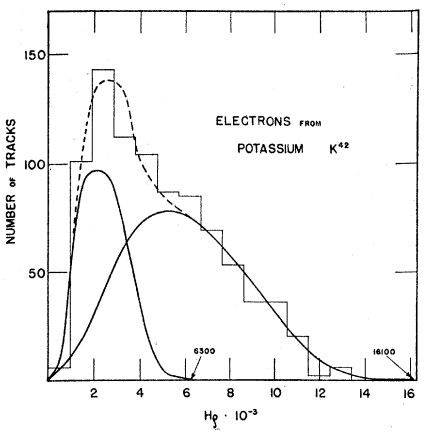

FIG. 11. Distribution histogram for the electrons from activated potassium (K^{42}). Here too the curve is too skew to be fitted by a single K–U curve but can be resolved into two as shown. The dotted line represents the sum of these two curves, and conforms very well to the histogram.

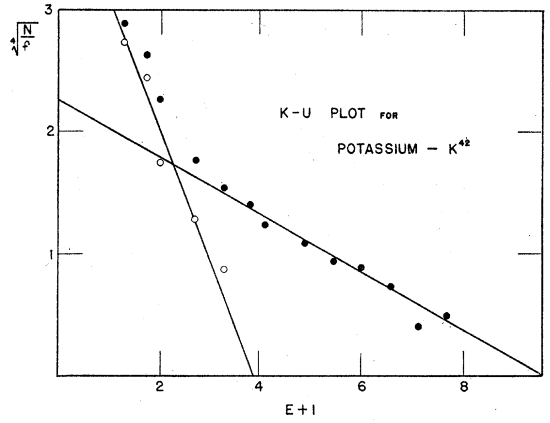

FIG. 12. K–U plot for activated potassium. This has been resolved into two lines by subtracting the "background" of the upper group to give the white points which then represent the lower group. The lines indicate upper limits for the two groups at $E+1=3.8$ and 9.6.

as yet succeeded in preparing sources of sufficient strength.

DISCUSSION

The data given above indicate that the Konopinski-Uhlenbeck theory gives a very good account of the shape of the distribution curves of the β-rays from the light radioactive elements. We have cited cases of three electron emitters (Na^{24}, Si^{31}, P^{32}) and two positron emitters (N^{13}, F^{17}) where deviations from the theoretical shape of the curve of the observed points are surprisingly small. The spectra of the three elements Cl, A^{41}, K^{42} can be resolved into two components each of which is very closely a K–U curve. This type of resolution is very similar to that proposed by Ellis and Mott[29] and does not seem to be completely unreasonable. The validity of such a resolution can perhaps only be tested when the energies and intensities of the γ-radiations from such bodies have been studied more closely, and also when more precise determinations of the β-ray endpoints have been made.

In fitting Eq. (4) to the data in any particular instance it is to be noted that although there are two constants appearing, only one is really adjustable, the other being determined by the number of tracks being considered. Also it is impossible to adjust the constant C in any way other than that we have used—any other adjustment, such as identifying it with the highest energy actually measured, will lead to a poorer fit between experiment and theory.

The high upper limits predicted by the K–U theory present a new difficulty in the study of the β-rays. If the order of contact with the axis is really as high as indicated by theory we must consider all the data hitherto gathered about upper limits as inexact. This would be very unfortunate, but the evidence seems strongly to indicate that it may be true. The very excellent agreement of the reaction energies in carbon found by Bonner, Delsasso, Fowler and Lauritsen and also by ourselves constitute the first direct information on this point. It may be that one must discount the fine correlation of the energies of the two branches of the Th C bodies obtained by Henderson[30] since it rests on two β-ray measurements whose upper limits are not greatly different. It is difficult to judge in this case since the authors give no experimental points on their curves by which one might estimate the chance that they could miss a tail with a high order of tangency to the momentum axis. Further evidence that such tails may really exist is to be found in the work of Scott mentioned above. Scott has carefully investigated the spectrum of Ra E and gives a higher upper limit than has previously been obtained. His data when plotted

[29] C. D. Ellis and N. F. Mott, Proc. Roy. Soc. **A141**, 502 (1933).
[30] W. J. Henderson, Proc. Roy. Soc. **A147**, 572 (1934).

in the form of Eq. (4) show the linear form very well and from the value of C indicated by them one deduces an upper limit very close to the one Scott gives.

Attempts have been made to fit the data of this paper on Sargent[31] curves. For the light radioactive bodies it would seem that one can at best say that no striking regularity is thus exposed.

The data presented above are summarized in Table I.

The authors are very much indebted to Professor E. O. Lawrence and Professor J. R. Oppenheimer for many discussions of this work.

[31] B. W. Sargent, Proc. Roy. Soc. **A139**, 659 (1933).

TABLE I. *Summary of values for the upper limit of β-ray spectra from various elements.*

SUBSTANCE	TARGET	UPPER LIMITS INSPECTION	*K–U* LINES
$N^{13}+$	Acheson graphite soot	1.3 MV	1.5 MV
$F^{17}+$	O_2	2.1	2.4
Na^{24}	NaF	1.7	1.95
Si^{31}	quartz	1.8	2.05
P^{32}	red P	1.8	2.15
Cl	AgCl	4.8	1.5 6.1
A	A gas	2.7	1.5∽5
K	KF	3.5	1.4, 4.4

We are also very grateful to the Research Corporation and the Chemical Foundation and the Josiah Macy, Jr. Foundation for grants whereby these experiments were made possible.

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The Ionization Gauge for Atomic Beam Measurements

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Use of two types of current measuring devices, galvanometer and vacuum tube amplifier, with an ionization gauge shows that the stable sensitivity,

$$\left(\frac{\text{positive ion current/mm gas pressure}}{\text{current fluctuation}}\right),$$

is greater for low electron emission densities thus favoring the use of the amplifier with its greater current sensitivity. Such factors as electrode size, relative potentials, electron emission, interelectrode leakage, type of inlet, gas adsorption, surface charges, temperature changes, B.K. oscillations, and variations of pumping speed which influence the design of a gauge are discussed and an arrangement to fulfill the necessary requirements is described. Performance tests show the gauge to be capable of measuring beam pressure changes at least as small as 3×10^{-11} mm of mercury or approximately 3×10^{9} mercury atoms per square centimeter per second.

THE properties of the ionization gauge method of measuring atomic beams are such that it would enjoy a wider field of application if the sensitivity were sufficient. Use by Zahl and Ellett[1] has shown no indication of the limitation attributed to adsorption phenomena by Johnson[2] but rather has indicated that the gauge might be capable of measuring beam pressure[3] changes of less than 10^{-10} mm of mercury. This, if true, would extend the range of usefulness to such a degree that a more careful investigation seemed advisable.

ANALYSIS OF MEANS OF EXTENDING SENSITIVITY

In normal use the sensitivity of an ionization gauge is determined by two factors, the magnitude of the electron current and the sensitivity of the positive ion current measuring instrument. Thus, to improve sensitivity, one may increase either the electron current or the sensi-

[1] Zahl and Ellett, Phys. Rev. **38**, 977 (1931).
[2] T. H. Johnson, Phys. Rev. **31**, 103 (1928).
[3] Unless otherwise indicated, all references to sensitivity are based on the effective pressure of the beam, expressed in millimeters of mercury, at the gauge inlet. Effective beam pressure is defined as that pressure inside a closed vessel which makes the number of molecules striking unit area of bounding surface in one second equal to the number transported by the beam across unit normal area in an equal time.