Short-Lived Radioactivities Induced in Fluorine, Sodium and Magnesium by High Energy Protons

M. G. WHITE, L. A. DELSASSO, J. G. FOX, AND E. C. CREUTZ
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey

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On the basis of simple theoretical considerations it is expected that short-lived positron emitters should be produced in fluorine, sodium, and magnesium under bombardment with 6-Mev protons. These activities have been found and ascribed to Ne^{19}, Mg^{23}, and $Al^{25, 26}$. Half-lives, positron spectra and threshold investigations have been made and found to compare very well with the theory. It is concluded that for isobars of the type $(n-p)=\pm 1$ the difference in binding energy is due solely to the effect of Coulomb forces up to at least mass number 25. The half-lives are found to depend on the inverse fifth power of the upper limit of the positron spectra in agreement with the theory.

I. INTRODUCTION

THE theoretical determination of nuclear binding energies is at present difficult because little is known about the exact character of the forces involved, and because of the many-body nature of the problem. By assuming equality of $n-n$ and $p-p$ interactions there results a great simplification in the calculations. In particular, for isobars formed by the interchange of all the protons and neutrons we are led to expect that almost[1] the entire difference in binding energy is due to Coulomb forces alone.[2] The assumption that neutrons and protons interact in the same way has been amply verified by recent binding energy calculations.[3] Several authors[4] have made binding energy calculations on this basis for isobars of the type $_4Be^7-{}_3Li^7$, $_5B^9-{}_4Be^9$, $_6C^{11}-{}_5B^{11}$, etc., which are characterized by $(n-p)=\pm 1$, i.e., the number of protons and neutrons differ by one. Positron emission by the heavier member of the pair then results in an interchange in the number of protons and neutrons, and the difference in binding energy may be attributed to Coulomb forces.

[1] A small correction arises from the slightly higher kinetic energy of the particles in the lighter member of the isobaric pair.
[2] W. Heisenberg, Zeits. f. Physik **77**, 1 (1932).
[3] E. Feenberg and E. Wigner, Phys. Rev. **51**, 95 (1937); W. Barkas, Phys. Rev. **55**, 691 (1939).
[4] H. A. Bethe, Phys. Rev. **54**, 436 (1938); E. Feenberg and J. Knipp, Phys. Rev. **48**, 906 (1935); S. Share, Phys. Rev. **50**, 488 (1936); H. Brown and D. Inglis, Phys. Rev. **55**, 1182 (1939); W. A. Fowler, L. A. Delsasso and C. C. Lauritsen, Phys. Rev. **49**, 561 (1936).

It is possible to calculate the classical electrostatic difference in energy if we make the simplifying assumption of a uniform distribution of charge throughout a volume which is proportional to the mass number. Barkas,[5] using Wigner's[6] formula, has determined the empirical value of the constant necessary to express the Coulomb energy as a function of the mass number. According to this work the Coulomb energy is given by $\Delta E=0.594(A-1)A^{-\frac{1}{3}}$ Mev where A is the mass number.

Comparison with experiment may be made by observing the upper limit of positron spectra of isobars of the above type, for the difference in binding energy is given by

$$\Delta E=E_{\max}{}^{+}+2e+(n-H).$$

Here $E_{\max}{}^{+}$ is the upper limit of the positron spectrum, and e, n, H are the masses of the electron, neutron and hydrogen atom, respectively. If positron emission always leads to an excited state it is of course necessary to add this excitation energy to $E_{\max}{}^{+}$.

Data prior to this paper have been thoroughly examined by various authors[7] who concluded that up to F^{17} the agreement between theory and experiment is surprisingly good. However, Si^{27}, formed by $Mg^{24}(\alpha, n)Si^{27}$, has a reported upper limit which ranges from 2 to 2.7 Mev and the expected upper limit is nearly 3.4 Mev. Between F^{17} and Si^{27} there were no reported activities

[5] W. Barkas, Phys. Rev. **55**, 694 (1939).
[6] E. Wigner, Phys. Rev. **51**, 947 (1937).
[7] See references 3 and 4 in particular.

although Ne^{19}, Na^{21}, Mg^{23}, and Al^{25} should be easily formed by p, n reactions if the bombarding energy exceeds the reaction threshold.[8] It is clearly of importance to fill up this gap in order to determine where these simple and apparently reasonable considerations begin to break down, if indeed they do. Assuming the applicability of the electrostatic energy formula we are led to expect an upper limit of 2.20, 2.79, and 3.07 Mev for Ne^{19}, Mg^{23}, and Al^{25}, respectively.

Nordheim and Yost[9] first pointed out that the absolute magnitude of the half-life could be predicted on the basis of the Fermi theory of β-decay. If one assumes that the positron transition is between ground states it is found that the half-life varies as $(E_{\max}^{+})^{-5}$. By extrapolation from F^{17}, O^{15}, N^{13}, C^{11} we expect the Ne^{19}, Mg^{23}, Al^{25} half-lives to be about 40, 12 and 8 sec., respectively. Wigner[10] has considered in detail the influence on the half-life of the possible transitions to excited states. He finds that for nuclei of this type the simple E^{-5} power dependence gives a fairly accurate representation of the data.

Production of these nuclei by $F^{19}(p, n)Ne^{19}$, $Na^{23}(p, n)Mg^{23}$, $Mg^{25}(p, n)Al^{25}$ reactions can take place only if the bombarding energy exceeds the value given by

$$E_p=(E_{\max}^{+}+2e+n-H)\frac{(A+1)}{A},$$

where E_p is the bombarding energy, A is the atomic weight of the bombarded nucleus and the factor $(A+1)/A$ takes care of recoil. We have again assumed that $E_{\max}^{+}$ corresponds to ground state transitions. The expected thresholds for these reactions are then 4.18, 4.78 and 5.06 Mev in the order given above. It is to be noted that threshold determinations give an independent measurement of the binding energy difference.[11]

We have looked for and found the expected activities. The remainder of this paper will be devoted to a discussion of the experimental data and their interpretation in the light of these considerations.

II. Experimental Procedure

Targets of compounds or pure elements were bombarded by protons with a maximum energy of 6.0 Mev and the resulting activity observed with a Lauritsen electroscope. Because of the short half-lives of the activities under investigation it was necessary to bombard outside the cyclotron so that targets could be quickly removed to the electroscope. An atmosphere of hydrogen surrounding the target was found important in order to avoid strong contamination activities arising from oxygen and nitrogen[12] in the air. To facilitate rapid measurements a moving tape chronograph was actuated by the observer as the fiber shadow moved across the divided scale. Observations could be started within 5 seconds after cessation of bombardment. Electroscope characteristics such as nonlinearity, lack of saturation at high ion densities, and polarization of the amber insulating post were investigated and corrected for in the final results. With a standard uranium β-ray source it was found that 1.2×10^4 disintegrations were required to produce a drift of one division.

Positron spectra and upper limits were secured by photographing tracks in a hydrogen-filled Wilson chamber situated in a magnetic field of 600 gauss constant to one percent over the chamber area. Although considerable care was exercised in measuring the field it is probably not accurate to better than one percent in absolute magnitude. Positrons entered the chamber through a 0.002″ copper window which formed part of a brass well let into the chamber top. In order to obtain several expansions per target bombardment, and still avoid an excess of tracks in the early expansions, a variable aperture lead slit was incorporated in the chamber well.

[8] Professor L. A. DuBridge has informed the authors that the Rochester group has produced a short period in sodium by proton bombardment.

[9] L. W. Nordheim and F. L. Yost, Phys. Rev. **51**, 942 (1937).

[10] E. Wigner, Phys. Rev. this issue.

[11] Even in the event that positron decay is always to an excited state the threshold determination provides a value of the binding energy difference. In view of the difficulty of securing gamma-ray energies the threshold measurement provides the most precise method of getting binding energy differences.

[12] Nitrogen has been investigated in this laboratory and found to yield a strong 20-min. C^{11} activity which is ascribed to $N^{14}(p, \alpha)C^{11}$. See W. Barkas, Phys. Rev. **56**, 287 (1939). This activity undoubtedly explains the 20-min. period earlier found in magnesium and reported by Ridenour, Delsasso, White and Sherr in Phys. Rev. **53**, 770 (1938).

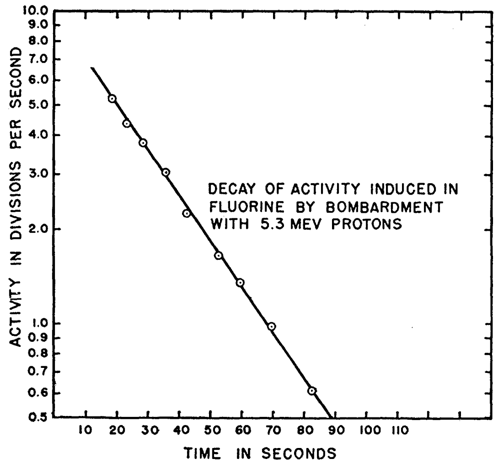

FIG. 1. Decay curve of Ne^{19}. Half-life 20.3±0.5 sec.

Measurement of track curvature was carried out by the customary method of reprojection. Care was taken to exclude from the measurements all scattered tracks and tracks too short to permit accurate curvature determinations.

III. Activities Investigated

Ne^{19}

Half-life.—We have already briefly reported[13] on the production of Ne^{19} by proton bombardment of fluorine compounds. Targets were prepared by melting a layer of lead fluoride on a backing strip of tantalum; lead and tantalum having previously shown no appreciable activity under bombardment with protons of this energy. In Fig. 1 is shown a typical decay curve for the radioactive neon resulting from a 1-min. bombardment of PbF_2. There is apparently only a single period and the mean value of several runs was found to be 20.3±0.5 sec.

Absorption measurements.—An attempt was made to secure the absorption curve of the emitted particles by using two electroscopes simultaneously. One electroscope was arranged to measure particles emerging from the back side of the target, and the other electroscope measured the activity from the front side after the particles had passed through a known thickness of aluminum. The ratio of these two observations at various absorber thicknesses is then a measure of the relative number of particles able to penetrate the aluminum absorbers. Since the first electroscope served merely as a monitor it was not necessary to know anything about the target thickness nor the stopping power of the tantalum. Complete decay curves were taken for every absorber and frequent checks were made on the relative efficiencies of the two electroscopes. Fig. 2 shows the resulting absorption curve, and though no precision is claimed it is clear that two conclusions may be drawn. First, that from Feather's[14] most recent mass-range expression the energy of the particles is about 2.3 Mev, and second, that there are no detectable gamma-rays except the annihilation radiation from the positrons. The latter conclusion may be inferred from the fact that after all the positrons were absorbed out there still remained a background of about 1.8 percent. This amount of residual ionization is interpreted as due to the positron annihilation radiation, for in the case of Zn^{63} about the same background was ob-

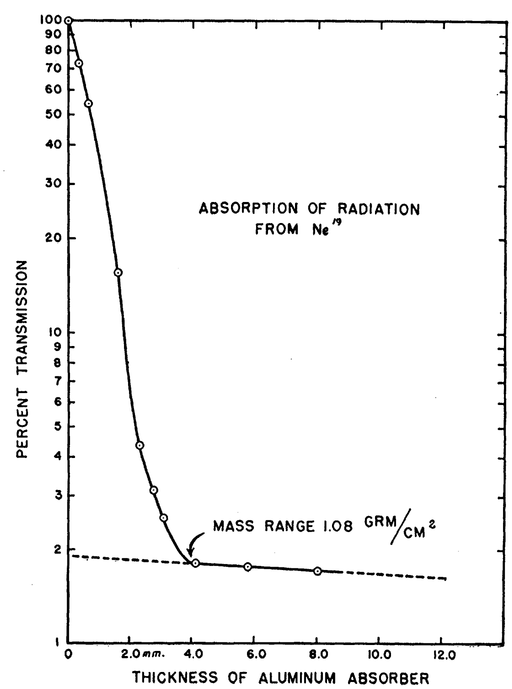

FIG. 2. Absorption curve of positrons and annihilation radiation from Ne^{19}. Upper limit of positrons as indicated by mass range is about 2.3 Mev.

[13] J. G. Fox, E. C. Creutz, M. G. White, L. A. Delsasso, Phys. Rev. **55**, 1106 (1939).

[14] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).

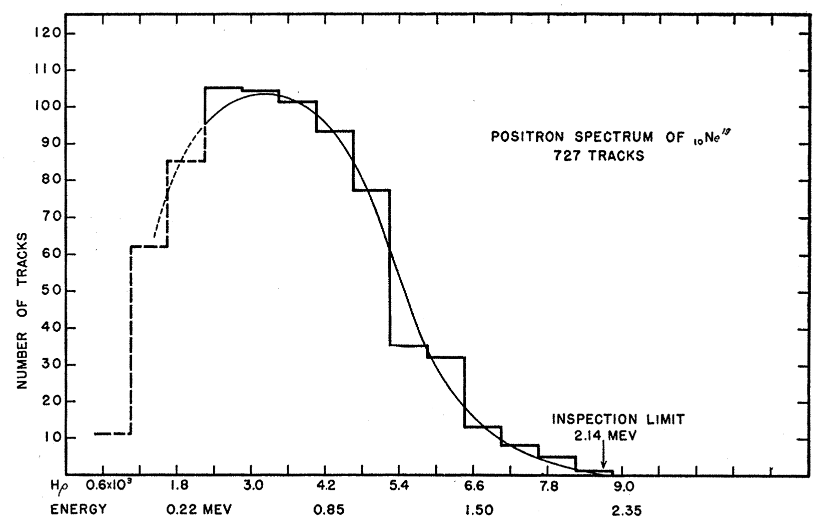

FIG. 3. Positron spectrum of Ne^{19}. Upper limit after foil correction is 2.20 Mev.

tained[15] and it was possible to show that the slope of the gamma-ray absorption curve corresponded to the expected annihilation energy. Obviously it is not possible to assert the complete absence of gamma-rays, but if *every* positron decayed to an excited state this would result in approximately 50 percent more background than that actually observed.

Positron spectrum.—Absorption measurements of electron energies are, in general, not reliable, for the degree of accuracy obtained depends on very careful observations near the endpoint where the intensity is low. Feather[14] has described a procedure for improving the determination of the mass range, but it was felt that more reliance could be placed in a cloud chamber investigation of the spectrum. We accordingly took about one thousand cloud chamber pictures with the resulting spectrum shown in Fig. 3. To reduce the chance of building up a long period contamination activity a large number of targets was employed. Although the shape of the spectrum is of considerable interest to the theory of β-decay we were chiefly concerned with the high energy end, and consequently the low energy portion was not accurately determined. Dotted lines indicate the regions of doubtful precision. By inspection the upper limit of the Ne^{19} spectrum was found to be 2.14 Mev, to which must be added 0.06 Mev lost in the copper window. The final upper limit of 2.20 Mev is thus seen to be in excellent accord with the expected value of 2.2 Mev.

In all this discussion we have assumed that the activity can only be ascribed to the production of Ne^{19}, for all other reactions involving the emission of heavy particles or gamma-rays lead either to stable isotopes or known activities of quite different periods.

Long periods in fluorine.—A one-hour bombardment of NaF with one microampere of 4.5-Mev protons gave weak 10-min. and 106-min. periods which are very probably contamination activities arising from traces of oxygen and carbon in the target. No attempt has been made to verify chemically this assumption.

Threshold determination.—Under ideal circumstances of a monochromatic proton beam and thin targets the measurement of the reaction threshold should provide a very accurate means of determining the difference in binding energy. However, the large spread in beam energy due to the cyclotron makes such an investigation difficult. A rough excitation curve was obtained by placing a PbF_2 target in the proton beam at various points along its path in air. After bombarding for a known time the target was removed and the activity measured with an electroscope. The curve of activity *versus* energy showed the steep rise characteristic of p, n

[15] L. A. Delsasso, L. N. Ridenour, R. Sherr and M. G. White, Phys. Rev. **55**, 113 (1939).

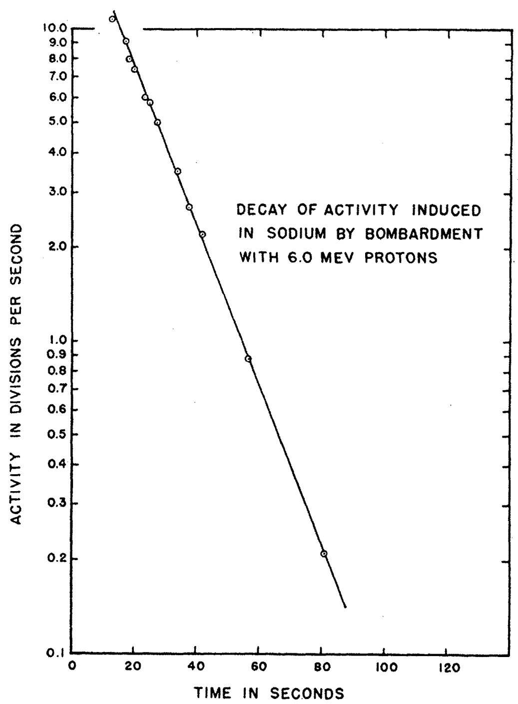

FIG. 4. Decay curve of Mg^{23}. Half-life 11.6 ± 0.3 sec.

reactions and the threshold was within ±0.25 Mev of the expected value of 4.18 Mev.

Mg^{23}

Half-life.—A short description of Mg^{23} was given at the Princeton meeting of the Physical Society.[16] Since that time more data have been collected and more accurate values for the upper limit and half-life obtained. Sodium, in the form of NaCl, was bombarded for one minute with 6.0-Mev protons. A characteristic decay curve is shown in Fig. 4. The half-life, as found by averaging several runs, is 11.6 ± 0.5 sec. We assume that the activity indicates the production of Mg^{23}, for all other possible reactions lead either to stable isotopes or well-known long periods. In a separate experiment $PbCl_2$ was found to yield no short periods when bombarded under the same conditions as the NaCl.

Positron spectrum.—In order to obtain the spectrum it was necessary to make over one thousand bombardments. To minimize the building up of long period contamination activities the targets were carefully scraped before each fresh bombardment. In Fig. 5 is shown the resulting spectrum. An upper limit of 2.82 Mev was found after correcting for absorption in the copper window. This is seen to agree very well indeed with the expected value of 2.79 Mev. As in the case of Ne^{19} we were largely concerned with the upper limit and consequently little attention was paid to the lower part of the spectrum.

Gamma-rays.—It is important to investigate the possible presence of gamma-rays before

[16] E. C. Creutz, M. G. White, L. A. Delsasso and J. G. Fox, Phys. Rev. **56**, 207(A) (1939).

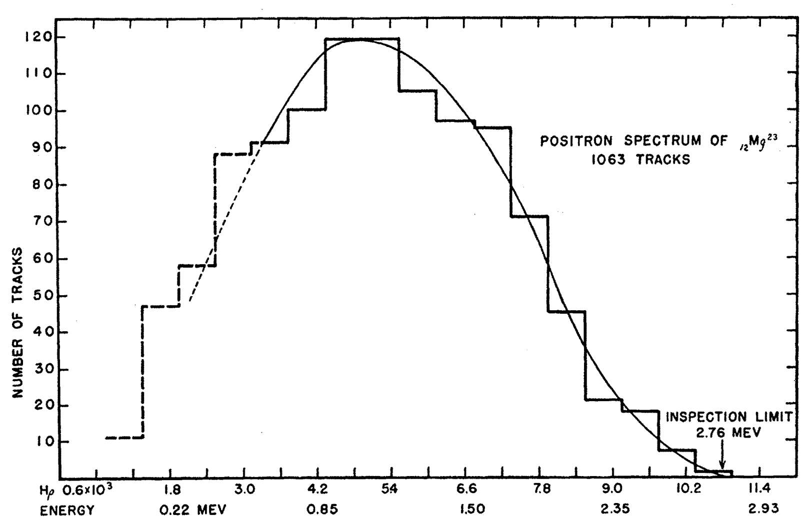

FIG. 5. Positron spectrum of Mg^{23}. Upper limit 2.82 Mev after foil correction.

assuming that the upper limit of the positron spectrum actually gives the difference in binding energy. A complete absorption curve would greatly aid in this decision, but unfortunately the short half-life makes such measurements difficult. We have accordingly limited the absorption measurements to just one thickness of absorber, the technique employed being the same as that described for Ne^{19}. When 1.25 cm of aluminum was interposed between target and electroscope the transmitted activity was found to be only 1.8 percent, from which we conclude, as in the case of Ne^{19}, that there are no detectable gamma-rays other than the annihilation radiation.

Threshold determination.—Difficulties attendant on securing the dependence of activity on bombarding energy have already been discussed for Ne^{19}. The shorter half-life of the radioactive magnesium makes such measurements even more troublesome; so the best that could be done was to insert aluminum foils in the path of the protons until no more activity was observed. We concluded that the activity disappeared at a proton energy within ± 0.3 Mev of the expected threshold of 4.78 Mev.

Long periods.—A search for long periods gave a negative result. We bombarded sodium metal in vacuum for 20 min. with 0.4 microampere of 5.5-Mev protons and found a fairly strong 20-min. activity. Further work with NaCl and nitrogen has convinced us that this period was due to $N^{14}(p, \alpha)C^{11}$ which arose from the nitrogen taken up by the sodium metal.

$Al^{25, 26}$

Half-life.—The study of Al^{25} which may be formed by the reaction $Mg^{25}(p, n)Al^{25}$ is complicated by the possibility of forming Al^{26} in the reaction $Mg^{26}(p, n)Al^{26}$. Frisch[17] was the first to report the production of Al^{26} by the reaction $Na^{23}(\alpha, n)Al^{26}$ and gave the half-life as 7 sec. Since sodium has but one isotope there is little doubt that his assignment is correct, especially in view of the detection of the emitted neutrons by Savel.[18] We have bombarded magnesium metal of commercial purity with 6.0-Mev protons and obtained the decay curve shown in Fig. 6. From the average of several runs we find the half-life to be 7.0 ± 0.5 sec. Although the decay curve seems to be simple, it cannot be determined whether two periods of nearly the same value are actually present, and it is impossible definitely to assign the activity.

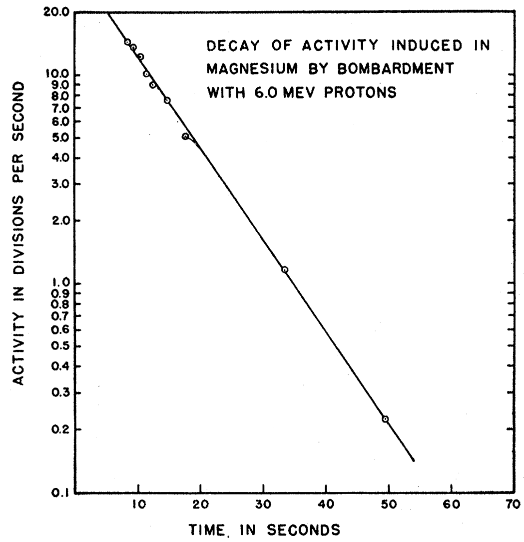

FIG. 6. Decay curve of activity induced in magnesium metal by 1-min. bombardment with protons of 6.0-Mev energy.

Positron spectrum.—Frisch[17] and Brandt,[19] using the absorption method, determined the upper limit of Al^{26} to be 1.8 and 1.5 Mev, respectively. As this value is far from the expected Al^{25} value of 3.07 Mev it was hoped that a study of the positron spectrum arising from our 7-sec. activity would help decide the question of isotope assignment. From Fig. 7 it is seen that the upper limit of the 7-sec. period induced by protons on magnesium is 2.99 Mev. This value compares very favorably with the expected Al^{25} upper limit of 3.07 Mev, and is in strong disagreement with the Al^{26} limit quoted above. Unfortunately little reliance is to be placed on absorption determinations; so we can not exclude the possibility that a large fraction of the spectrum near the upper end is due to Al^{26}. If one takes the absorption measurements at their face value it is clear that the high energy end of Fig. 7 can be ascribed to Al^{25}, and the low energy end to Al^{26}. Difficulties in measuring low energy positrons with a magnetic field adjusted for the high energy tail make uncertain any attempted

[17] O. Frisch, Nature **133**, 721 (1934).
[18] M. P. Savel, Comptes rendus **198**, 1404 (1934).
[19] H. Brandt, Zeits. f. Physik **108**, 726 (1938).

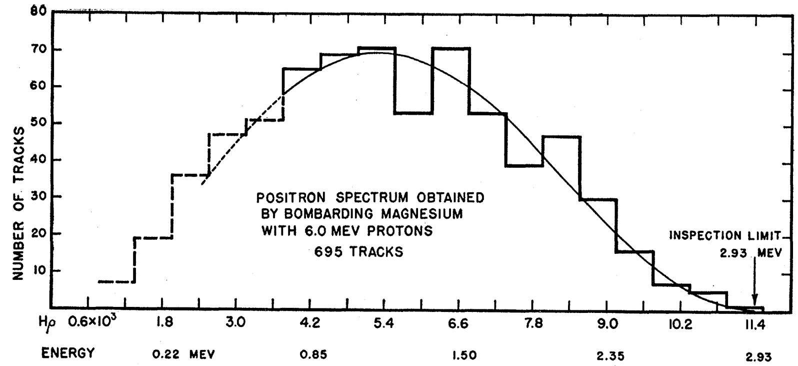

FIG. 7. Positron spectrum of activity induced in magnesium metal by 6.0-Mev protons. Upper limit after foil correction is 2.99 Mev.

resolution of the curve into two components. Nevertheless the shape of the curve does not indicate a decided preponderance of low energy positrons.

Threshold.—On theoretical grounds[3, 4] it is quite certain that the $Mg^{26}-Al^{26}$ binding energy difference is only a little less than that for $Mg^{25}-Al^{25}$, and there seems no reason to suppose that both activities are not present in the decay curve and in the spectrum. On the other hand, Al^{24} certainly cannot be formed at our energies from Mg^{24} if the threshold is actually 13 Mev as deduced from Barkas' table of computed masses. Actual threshold measurements indicate that there is no detectable activity below 5 Mev, in agreement with the expected Al^{26} threshold of about 4.5–5 Mev, and we must therefore conclude that the reported upper limit of Al^{26} is in error. Another alternative is to assume that Al^{26} always decays to an excited state about 1.2 Mev above the ground level. This would, however, not be in accord with the short half-life.

IV. DISCUSSION

Our measurements indicate that up to mass 23 and possibly even 25 the simple picture discussed at the beginning of this paper is able to account for the binding energy differences of isobars of the type $(n-p)=\pm 1$. Deviations might have been expected if shell structure plays an important role in determining the size of the nucleus, but apparently such effects are not important. The further conclusion may be drawn that there is no appreciable swelling of the nucleus because of Coulomb forces. The work also gives additional support to the hypothesis of equal $n-n$ and $p-p$ interactions.

From the half-life measurements and their comparison with the values as predicted on the basis of the Fermi theory it seems evident that in general for light elements the inverse fifth power dependence of half-life on energy gives a reasonably good description of the facts.

V. ACKNOWLEDGMENTS

It is a pleasure to thank Professor Wigner for pointing out the interest in this problem and for many discussions on the theoretical questions involved. The assistance of Dr. Barkas in making some of the measurements is also appreciated.