may proceed by emission of tritons are given in Table II. The Cu^{65} targets did show a yield of Cu^{62} below 25 Mev, but this can be largely accounted for by the Cu^{63} contamination. The maximum yield of the (p, H^3n) reaction in this region is of the order of 1 millibarn. The Cu^{63} targets have a cross section of 1.2 millibarns for the production of Cu^{61} at 15 Mev. The energy uncertainty here is ± 2.5 Mev so this may be partly due to the (p, dn) reaction. At 10 Mev the cross section is about 0.1 millibarn and should be entirely due to the (p, H^3) reaction.

The author wishes to thank Mrs. A. M. Dean and Miss Margaret Heineman for their assistance in performing these experiments and Mr. Richard C. Wharton and the cyclotron crew for their cooperation during the bombardments. Dr. Richard Diamond and Dr. Karl Strauch are responsible for some very helpful discussion and criticism in preparing this report.

PHYSICAL REVIEW VOLUME 91, NUMBER 4 AUGUST 15, 1953

The Radiations of U^{240} and Np^{240}†

J. D. KNIGHT, M. E. BUNKER, B. WARREN, AND J. W. STARNER
Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico

(Received April 21, 1953)

The beta and gamma radiations of U^{240} and Np^{240} have been examined with magnetic lens and scintillation spectrometers. $U^{240}(t_{\frac{1}{2}}=14.1\pm0.2$ hours) emits a single beta-ray group of maximum energy 0.36 Mev. No gamma rays were observed. $Np^{240}(t_{\frac{1}{2}}=7.3\pm0.3$ minutes) emits four beta-ray groups with maximum energies of 2.156, 1.59, 1.26, and 0.76 Mev and relative intensities of 52, 31, 11, and 5.4 percent, respectively. Gamma rays having energies of 1.40, 0.90, and 0.56 Mev were observed. These data can be fitted into a simple decay scheme. On the basis of cycles involving the total decay energies of these isotopes, plus published data on alpha- and beta-transition energies and photoneutron thresholds of related nuclides, the binding energy of the last neutron in U^{240} is calculated to be 5.92 ± 0.15 Mev, and that of the last neutron in Np^{240} to be 4.98 ± 0.15 Mev.

INTRODUCTION

THE isotopes U^{240} and Np^{240} were found by Hyde and Studier[1] in uranium which had been irradiated at high neutron flux in a pile, the U^{240} having been produced by the neutron capture sequence $U^{238}(n,\gamma)U^{239}(n,\gamma)U^{240}$. The U^{240} and its daughter Np^{240}, both found to be beta emitters, were identified by standard radiochemical techniques. The half-lives were reported as 14 ± 2 hours and 7.3 ± 0.3 minutes, respectively. A later report[2] from the same laboratory gave a revised U^{240} half-life of 17 hours. Further characterization of these isotopes was limited by the low attainable specific activity of the U^{240} and by the presence of a relatively large amount of U^{237} which was also produced during the irradiation by the reaction $U^{238}(n,2n)U^{237}$.

With the availability at this laboratory of high-neutron-flux devices and of the means of retrieving portions of target materials placed in or near them, it has been possible to obtain U^{240} sources of sufficient strength for beta-ray spectrometry and without serious U^{237} interference. This report describes the results of an investigation of the radiations of the U^{240}-Np^{240} chain, conducted on a series of such sources.

APPARATUS AND SOURCE PREPARATION

The beta counting for measurement of half-life and for verification of parent-daughter relationship of the observed uranium and neptunium activities was done with continuous-flow methane gas proportional counters. These counters have a dead-time loss of about 1 percent at 10^5 counts per minute so that sample decay can be followed through factors of the order of 10^4 in counting rate.

The beta-ray spectrum of the U^{240}-Np^{240} equilibrium mixture was measured with a magnetic lens spectrometer.[3] The baffles were adjusted to give a resolution of 3.5 percent. The electron detector was an end-window Geiger tube with a 0.5-in. diameter aperture and a 2.4 mg/cm² mica window having a low energy cutoff at about 31 kev.

The gamma-ray data were obtained with a scintillation spectrometer consisting of a NaI(Tl) crystal attached to an RCA type 5819 photomultiplier. A 1-in. aperture lead collimator was placed between the source and the crystal. Two different scintillation crystals were used: a large one, 2 in. in diameter and 2 in. high, for examination of high-energy gamma rays and general coverage of the entire spectrum, and a smaller one, about 0.25 in. thick, for more detailed examination of the low-energy portion of the spectrum. The pulses were sorted with a ten-channel analyzer.

The uranium activity was isolated from the irradiated

† Work done under the auspices of the U. S. Atomic Energy Commission.

[1] E. K. Hyde and M. H. Studier, Argonne National Laboratory Reports ANL-4143, April 15, 1948, and ANL-4182, August 4, 1948 (unpublished).

[2] Studier, Magnusson, Siddall, and Huizenga, Argonne National Laboratory Report ANL-4667, May 1, 1951 (unpublished).

[3] L. M. Langer, Phys. Rev. **77**, 50 (1950).

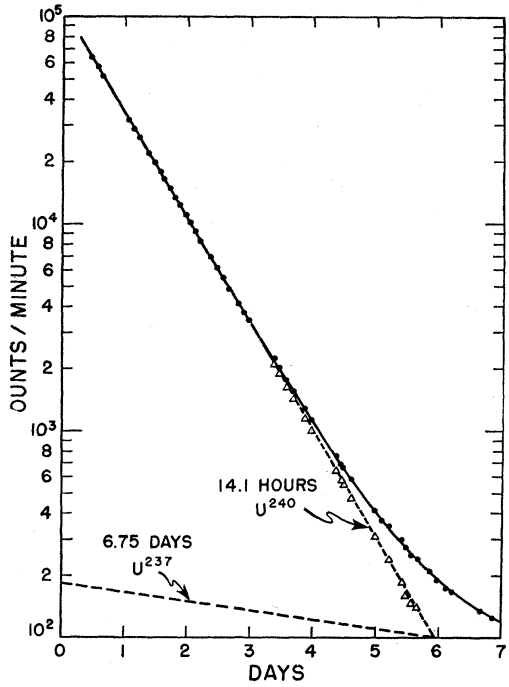

FIG. 1. Decay curve of U^{240}-Np^{240} in secular equilibrium. ●, original points; △, activity after subtraction of 6.75-day component.

source material, which contained Np^{239} and fission products as the principal beta-emitting contaminants,

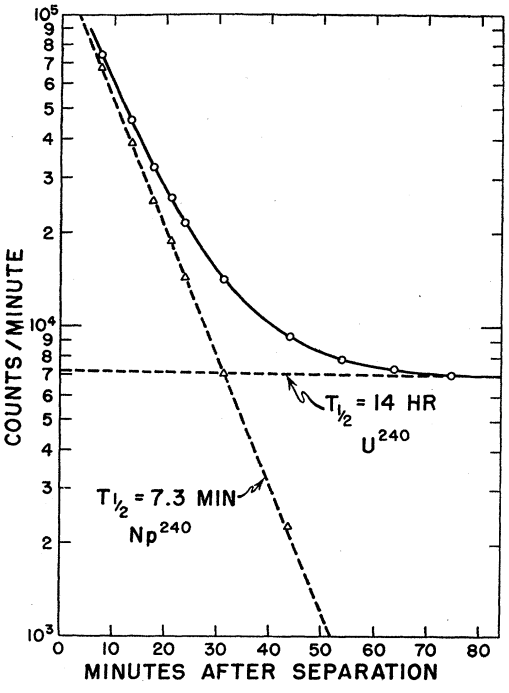

FIG. 2. Decay curve of Np^{240}. The 14-hour background is due to a trace of uranium carried along with the neptunium fraction in the rapid chemical separation. ○, original points; △, activity after subtraction of 14-hour component.

by a standard chemical analysis using uranium carrier and a series of product and by-product precipitations. Although the use of uranium carrier in the isolation procedure meant that the final sources were rather heavy for beta spectrometry (up to 1 mg), this approach was chosen in preference to a carrier-free extraction in the interests of reliable beta decontamination combined with good chemical yield.

For beta-ray spectrometry, the U^{240} source (with Np^{240} grown into secular equilibrium) was mounted as ~1 mg of $(NH_4)_2U_2O_7$, spread over an area of 1 cm^2 on a backing of rubber hydrochloride (0.5 mg/cm^2) and covered with a thin Zapon film (~0.02 mg/cm^2). Similar sources were used in most of the scintillation work. For scintillation examination of the Np^{240} alone, a series of neptunium fractions were isolated from the parent uranium solution by a rapid chemical procedure which involved complexing the uranium with NH_2OH and co-precipitating the neptunium with LaF_3.

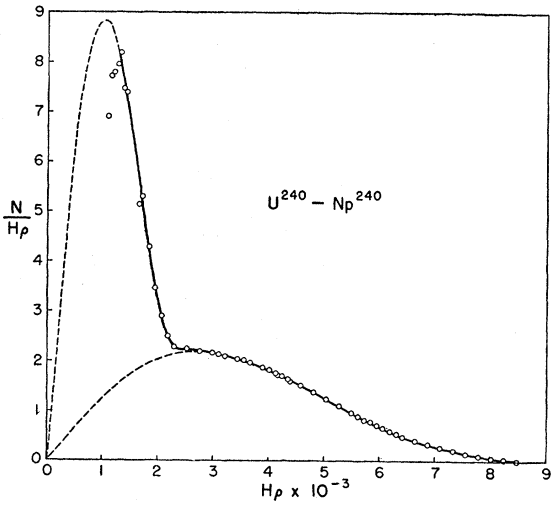

FIG. 3. Beta spectra of U^{240}-Np^{240} in secular equilibrium.

EXPERIMENTAL RESULTS

Typical beta-decay curves of the U^{240}-Np^{240} equilibrium mixture and of the neptunium fraction separated from the mixture, as followed on a beta proportional counter, are shown in Figs. 1 and 2, respectively. The equilibrium mixture was counted through 86 mg/cm^2 of aluminum absorber to suppress the contribution of the U^{237} activity unavoidably present in the sample. Under these conditions practically all of the U^{240} beta rays are suppressed also, and the bulk of the observed counts come from the hard beta rays of the neptunium daughter; however, because of the short half-life of Np^{240}, the observed rate of decay is that of the U^{240} parent. These measurements yielded a U^{240} half-life of 14.1±0.2 hours, and a Np^{240} half-life of 7.3±0.3 minutes.

Because of the contribution of U^{237} to the low-energy

regions of the beta and gamma spectra, it was necessary to make two sets of measurements on each source, the first as soon as possible after preparation of the source, when the U^{240}/U^{237} ratio was most favorable, and the second about a week later, when only the U^{237} remained. The U^{237} spectra were corrected back to the time of the original measurement and subtracted from the composite data to give the net spectra of the U^{240}-Np^{240} equilibrium mixture. The observed U^{240}-Np^{240} electron momentum distribution, corrected for U^{237}, is plotted in Fig. 3. The high-energy portion of the momentum distribution, which was found to belong to Np^{240}, is re-plotted in Fig. 4. A Fermi analysis of the data (Figs. 5 and 6) resolved the spectrum shown in Fig. 3 into five groups with end-point energies of 2.156±0.01, 1.59±0.01, 1.26±0.01, 0.76±0.02, and 0.36±0.02 Mev. The calculated relative intensities and log *ft* values of these groups are listed in Table I. The Fermi plot of

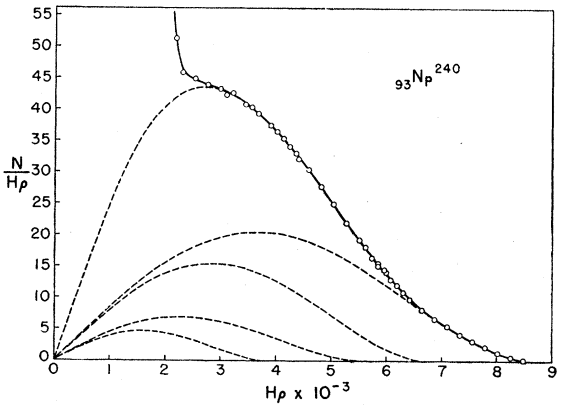

FIG. 4. Beta spectrum of Np^{240}. The partial spectra were determined from the Fermi plot analysis. The upturn below $H\rho = 2800$ is the start of the U^{240} spectrum.

the 0.36-Mev group (Fig. 6) exhibits a slight upward curvature. Such behavior is expected of a low-energy spectrum obtained from a source as thick as 1 mg/cm². The drop-off below $W \approx 1.25$ is due to counter-window absorption. The relative scarcity of experimental points on this group is a consequence of the interference of the U^{237} spectrum. The U^{237} conversion lines were so strong that data taking had to be confined to the valleys between the peaks.

The scintillation spectrum of the gamma rays of a sample containing U^{240}, Np^{240}, and U^{237}, taken with the 2-in. NaI(Tl) crystal, is plotted in Fig. 7. The lower curve shows the three prominent U^{237} peaks at 0.060, 0.096, and 0.206 Mev, plus a U^{240}-Np^{240} peak at 0.560 Mev. Data taken with the same crystal and phototube but with lower amplifier gain, shown in the inset, indicate additional peaks at 0.90 and 1.40 Mev. When the photopeak areas were corrected for crystal sensitivity, the relative intensities of the 0.56-, 0.90-, and 1.40-Mev gamma rays were found to be, respectively,

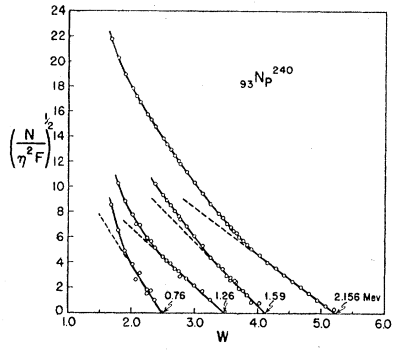

FIG. 5. Np^{240} Fermi plot. The break below $W = 2$ is due to U^{240}.

63:26:10. A similar examination of the radiations from the neptunium fraction, obtained by rapid milking from a solution of the U^{240}-Np^{240}-U^{237} mixture, revealed peaks at 0.56 and 0.90 Mev which decayed with a half-life of a few minutes. Counts were also observed above 1 Mev, but the sources were too weak to permit resolution of a meaningful peak (or peaks) in this energy region. Subtraction of the U^{237} background from the gamma-ray spectrum of the U^{240}-Np^{240}-U^{237} mixture revealed no evidence of gamma peaks from 0.5 Mev down to approximately 10 kev, the practical low-energy detection limit of the scintillation apparatus.

Following the observation of the three above-mentioned gamma rays, the beta spectrum was re-examined in an effort to find conversion lines corresponding to them. No conversion lines were found in the high-energy region, and none in the low-energy region which were not attributable to U^{237}. On the basis of counting statistics for the portions of the momentum spectrum involved, it is estimated that for each of these gamma rays, the number of conversion electrons per beta disintegration is less than 0.003.

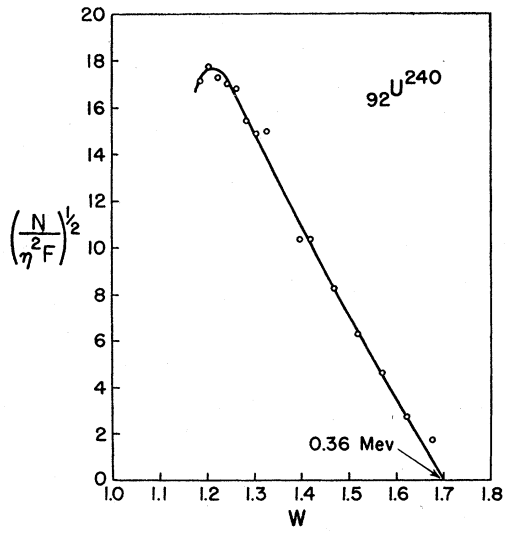

FIG. 6. U^{240} Fermi plot.

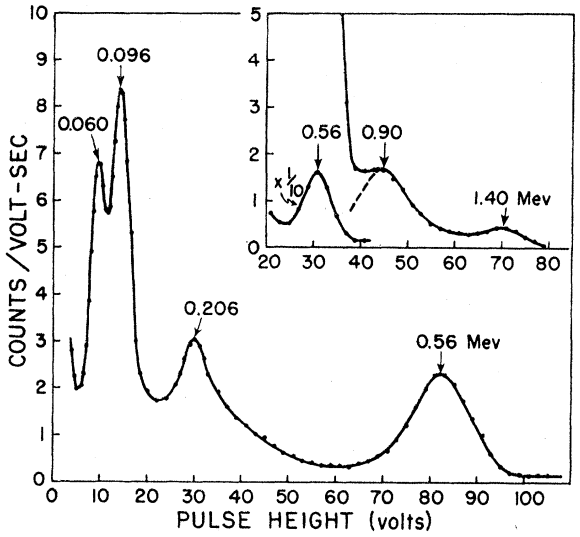

FIG. 7. Scintillation spectrum of U^{240}-Np^{240} in secular equilibrium, plus U^{237} contaminant. Peaks at 0.060, 0.096, and 0.206 Mev are due to U^{237}. Inset: high-energy region taken at lower amplifier gain.

DISCUSSION

Since beta-spectrum measurements were performed only on the U^{240}-Np^{240} equilibrium mixture, assignment of the beta components to the individual nuclides has to be made partly on the basis of other data. It was known from aluminum absorption measurements performed one the neptunium fraction separated from the equilibrium mixture that Np^{240} has a beta-ray end point of approximately 2 Mev. Therefore, the 2.156-Mev group almost certainly belongs to Np^{240}. Also, when 0.56 and 0.90 Mev, the energies of the gamma rays observed to belong to the neptunium fraction, are subtracted from the maximum beta-energy, the remainder energies correspond to the next two lower beta groups indicated by the Fermi analysis. By the same subtraction process, the 1.40-Mev gamma ray evidently follows the 0.76-Mev beta component. Thus, it appears that the four beta groups with end-point energies of 2.156, 1.59, 1.26, and 0.76 Mev all belong to Np^{240}. Furthermore, the fact that the sum of the intensities of these four groups, reconstructed from the Fermi plot analysis, is equal (within experimental error) to the intensity of the 0.36-Mev beta-ray group is a fairly strong argument that (1) the 0.36-Mev group belongs to U^{240}, and (2) the 0.36-Mev group is the only beta group associated with U^{240}. The assignment of only the 0.36-Mev beta group to U^{240} is supported also by the following negative evidence: if the intense 0.36-Mev transition led to an excited state of Np^{240}, a correspondingly strong gamma ray and/or x-rays resulting from its internal conversion would be emitted. Such a strong gamma ray could not have escaped detection, and, although the scintillation data indicate the presence of neptunium K and L x-rays, their intensity and decay rate suggest that they are due entirely to the U^{237} in the sources. Therefore, it appears that the 0.36-Mev beta group represents a ground-state transition, although the experimental data do not exclude the possibility that U^{240} decays to a level of Np^{240} which has a lower excitation energy than the L binding energy of neptunium, or $\sim$20 kev. The above conclusions are summarized in the form of the decay scheme shown in Fig. 8. It is possible that in the decay of both U^{240} and Np^{240}, there are weak radiations which went unobserved. For example, it is known from the decay of Cm^{244} that there is a 41-kev excited state in Pu^{240},[4] and there may be some branching to this level from the higher Pu^{240} levels which become excited through the decay of Np^{240}. Therefore, it is proposed only that the decay scheme of Fig. 8 represents the main decay branches of the two nuclides.

There is little that can be said about spin and parity assignments. The ground states of U^{240} and Pu^{240} are, of course, assumed to be (0,+). The log ft value of the 2.156-Mev beta group is 6.5, which suggests that this transition is once forbidden, $\Delta I = 0,1$. The assignment of odd parity to the ground state of Np^{240} is in agreement with shell-model predictions. One might expect the U^{240}-Np^{240} ground-state transition to have the same ft value as the Np^{240}-Pu^{240} ground-state transition, since identical spin and parity changes are involved. However, log ft of the U^{240}-Np^{240} transition is only 5.6. This apparent discrepancy is not considered to be a reliable indication of whether or not the 0.36-Mev beta group is a ground-state transition, particularly since similar ft anomalies are to be found in the literature, e.g., Ce^{144}-Pr^{144}-Nd^{144}.[5]

TABLE I. The beta-ray groups of U^{240} and Np^{240}.

End-point energy (Mev)	Relative intensity	log ft[a]	Isotopic assignment
2.156	52	6.5	Np^{240}
1.59	31	6.3	Np^{240}
1.26	11	6.3	Np^{240}
0.76	5.4	5.9	Np^{240}
0.36	104	5.6	U^{240}

[a] Determined from the curves given by S. A. Moszkowski, Phys. Rev. **82**, 35 (1951).

On the basis of the ground-state decay energies of 0.36 Mev and 2.156 Mev assigned to U^{240} and Np^{240}, respectively, it is possible to fit these nuclides into cycles from which their last-neutron binding energies may be obtained. The cycles and transition energies which were used are diagrammed in Fig. 9. From the cycle U^{239}-Np^{239}-Pu^{239}-Pu^{240}-Np^{240}-U^{240}, the binding energy of the last neutron in U^{240} is calculated to be 5.92 ± 0.15 Mev, and that of the last neutron in Np^{240} to be 4.98 ± 0.15 Mev. The last-neutron binding energy

[4] F. Asaro and I. Perlman, Phys. Rev. **87**, 393 (1952).
[5] F. T. Porter and C. S. Cook, Phys. Rev. **87**, 464 (1952).

of U^{240} can also be obtained, although with less accuracy, from the cycle U^{239}-U^{238}-U^{237}-U^{236}-Pu^{240}-Np^{240}-U^{240}. The value obtained in this manner is 6.32 Mev. The discrepancy between these two numbers is probably due to inaccuracy in the reported last-neutron binding energies of U^{237}, U^{238}, and U^{239}. Since the last-neutron binding

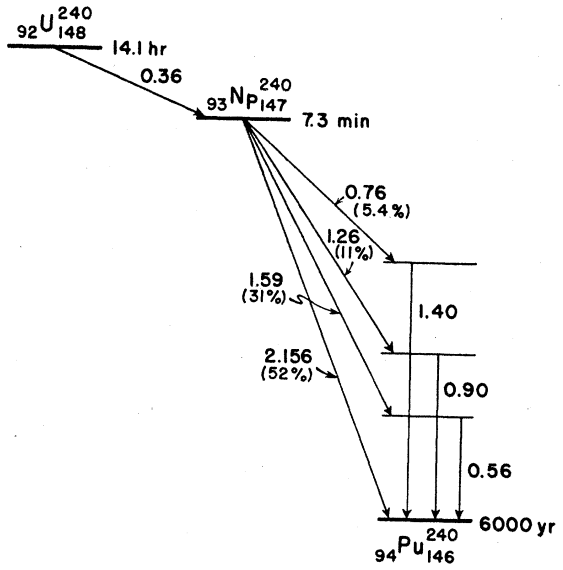

FIG. 8. Proposed decay scheme of U^{240}-Np^{240}.

energy of U^{240} might be expected to be several hundred kilovolts less than that of U^{238} (5.97 Mev), the value 6.32 Mev appears to be definitely out of line and even 5.92 Mev is disturbingly high.

A possible reason for the high value of the U^{240} one-neutron binding energy is that 148 neutrons form a

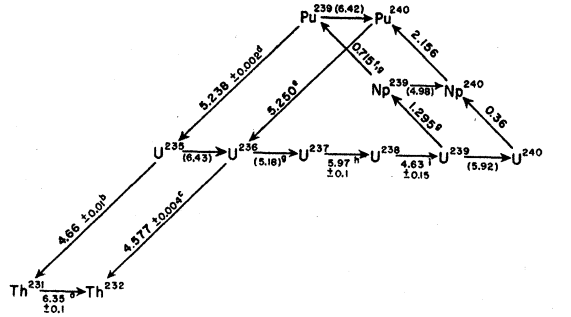

FIG. 9. Cycles used to determine the last-neutron binding energies of U^{240} and Np^{240}. Calculated values are in parentheses; directly observed values are not. References are as follows:

a—Magnusson, Huizenga, Fields, Studier, and Duffield, Phys. Rev. **84**, 166 (1951).
b—A. Ghiorso, Phys. Rev. **82**, 979 (1951).
c—Jaffey, Diamond, Hirsch, and Mech, Phys. Rev. **84**, 785 (1951).
d—F. Asaro and I. Perlman, Phys. Rev. **88**, 828 (1952).
e—F. Asaro and I. Perlman, cited by Hollander, Perlman, and Seaborg, University of California Radiation Laboratory Report UCRL-1928, August, 1952 (unpublished).
f—Tomlinson, Fulbright, and Howland, Phys. Rev. **83**, 223 (1951).
g—Huizenga, Magnusson, Freedman, and Wagner, Phys. Rev. **84**, 1264 (1951).
h—Huizenga, Magnusson, Fields, Studier, and Duffield, Phys. Rev. **82**, 561 (1951).
i—J. A. Harvey, Phys. Rev. **81**, 353 (1951).

closed shell. Dunlavey, in a study of nuclear radii calculated from alpha decay energies and alpha half-lives,[6] finds some indication for closed shell structure at Cm^{244}, a 148-neutron nucleus. Although there are few known nuclear spins on which to base definite assignments of single-particle levels in this region, an examination of the levels available beyond the 126-particle shell would suggest that a 148-particle shell could be achieved by the configuration

$$(1i_{11/2})^{12}(2g_{9/2})^{10}.$$

[6] D. C. Dunlavey, University of California Radiation Laboratory Report UCRL-1911, August, 1952 (unpublished).