

Energy Levels of Np^{237} Populated by the Beta Decay of U^{237}

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A spectroscopic investigation has been made of the radiations from U^{237} with magnetic 180° spectrographs and a double-focusing spectrometer, and with scintillation and coincidence spectrometers. Energy levels are identified at 0, 33.20, 59.57, 103.0, 267.5, 332.3, 368.5, and 370.9 keV, and spin and parity assignments are given for all the levels from the gamma-ray multipolarity and beta-decay information. Conversion-coefficient information is presented, and an anomaly in the electric-dipole conversion coefficients is discussed. A new rotational band with $K=1/2$ is postulated, with base state at 332.3 keV.

Correlations of the data with various aspects of the Bohr-Mottelson nuclear model are made, in the following categories: (1) energy-level spacings and moments of inertia; (2) gamma-ray relative intensities and the K -selection rules and branching-ratio rules; (3) correlations of spins and parities with the Nilsson numbers (N, n_z, Λ) of these states; (4) correlation of beta and gamma lifetimes with selection rules in the asymptotic quantum numbers.

INTRODUCTION

THE energy levels of Np^{237} are significant from the point of view of the Bohr-Mottelson nuclear model for spheroidal nuclei because of the appearance of two rotational bands near the ground state with quite different moments of inertia and magnetic moments.¹ Studies of these states populated by alpha decay of Am^{241} have been carried out by several investigators,¹⁻⁸ and a review of their properties has been given by Perlman and Rasmussen⁹ in their paper on alpha decay.

It has been demonstrated that such properties as moments of inertia and magnetic moments can be affected by the presence of higher-lying states of similar configuration.^{10,11} Hence, in order to form a more complete picture of the energy levels of Np^{237} , it is of interest to examine the higher excited states of Np^{237} which are populated by the beta-decay of U^{237} , states which are only very weakly excited by the alpha decay of Am^{241} and not at all by the electron-capture decay of Pu^{237} because of its low disintegration energy.^{12,13}

The study of U^{237} by Wagner *et al.*¹⁴ has indicated that the beta decay of this nuclide populates primarily an excited state of Np^{237} at 269 keV; their results have been confirmed and extended by the recent work on U^{237} by Baranov and Shlyagin¹⁵ in which multipolarity information about several of the gamma rays is presented. The present study had as its objectives a more precise measurement of the energies of the U^{237} gamma rays and the determination of the multiplicities, intensities, and coincidences of the gamma rays so that the previous comparisons^{1,16} of the properties of Np^{237} with predictions of current nuclear models could be extended to include the higher excited states. As a result of our studies several modifications of the earlier decay schemes seem necessary.

Subsequent to the first writing of the present paper there has come to our attention the detailed beta spectroscopic and coincidence study of U^{237} by Bunker, Mize, and Starner.¹⁷ We have not attempted to obtain and compare in detail their results except for the summary information appearing in an abstract.¹⁸ Their measurement of the lifetime of the 267.5-keV level as $5.4 \pm 0.5 \times 10^{-9}$ sec is a most important piece of new information. The chief similarities and differences in the recent studies will be made clear by the comparison of reported gamma transition energies in Table II.

PREPARATION OF SAMPLES

The U^{237} samples were prepared by one-day irradiations of approximately 100-microgram amounts of U^{238} in the Materials Testing Reactor at Arco, Idaho at a flux of $\sim 2 \times 10^{14}$ neutrons/cm² second.

The following chemical purification of the uranium was carried out: The target material was dissolved in

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¹ Hollander, Smith, and Rasmussen, *Phys. Rev.* **102**, 1372 (1956). This paper will hereafter be referred to as HSR.

² Jaffe, Passell, Browne, and Perlman, *Phys. Rev.* **97**, 142 (1955).

³ P. P. Day, *Phys. Rev.* **97**, 689 (1955).

⁴ Milsted, Rosenblum, and Valadares, *Compt. rend.* **239**, 259, 700 (1954).

⁵ S. A. Baranov and K. N. Shlyagin, *Conference of the Academy of Sciences of the U.S.S.R. on the Peaceful Uses of Atomic Energy, July 1-5, 1955* (Akademiia Nauk, S.S.S.R., Moscow, 1955), translated by Consultants Bureau, New York, 1955, Vol. 1, p. 183.

⁶ J. O. Newton, *Nature* **175**, 1028 (1955); and private communication, October, 1955.

⁷ Beling, Newton, and Rose, *Phys. Rev.* **87**, 670 (1952).

⁸ Krohn, Novey, and Raboy, *Phys. Rev.* **98**, 1187 (1955).

⁹ I. Perlman and J. O. Rasmussen, *Handbuch der Physik* [Springer-Verlag, Berlin (to be published)], Vol. 42.

¹⁰ A. K. Kerman, *Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd.* **30**, No. 15 (1956).

¹¹ R. J. Blin-Stoyle and M. A. Perks, *Proc. Phys. Soc. (London)* **A67**, 885 (1954).

¹² Glass, Thompson, and Seaborg, *J. Inorg. Nuclear Chem.* **1**, 3 (1955).

¹³ R. W. Hoff and M. I. Kalkstein (unpublished results).

¹⁴ Wagner, Freedman, Engelkemeir, and Huizenga, *Phys. Rev.* **89**, 502 (1953).

¹⁵ S. A. Baranov and K. N. Shlyagin, *J. Exptl. Theoret. Phys.* **30**, 225 (1956) [translation: *Soviet Phys. JETP* **3**, 200 (1956)].

¹⁶ J. O. Rasmussen, *Arkiv Fysik* **7**, 185 (1953).

¹⁷ M. E. Bunker (private communication, January, 1957).

¹⁸ Bunker, Mize, and Starner, *Bull. Am. Phys. Soc. Ser. II*, **2**, 104 (1957), and verbal report.

TABLE I. Conversion electron data for U^{237} .^a

| Electron energy (keV) ^b | Shell ^c | Transition energy (keV) | Instrument ^d quoted | Visual intensity estimates ^e | | | Intensity (densitometer) | Intensity (double-focusing) | Remarks |
|------------------------------------|--------------------|-------------------------|--------------------------------|---|-------------------|--------------------|--------------------------|--|---------|
| | | | | PM I (plate 264) | PM II (plate 272) | PM III (plate 279) | | | |
| 20.64 | M_I | 26.38 | I | w | | | | | |
| 21.05 | M_{II} | ~26.4 | I | m | | | | Composite with 43.5 L_I line | |
| 21.94 | M_{III} | 26.36 | I | vw | | | | Not well resolved from 43.5 L_{II} line | |
| 22.44 | M_{IV} | 26.29 | I | vw | | | | | |
| 24.86 | N_I | 26.36 | I | w | | | | | |
| 25.02 | N_{II} | 26.34 | I | wm | | | | N_{III} slightly less intense than N_I | |
| 25.28 | N_{III} | 26.36 | I | w | | | | | |
| 26.11 | O | ~26.4 | I | vvw | | | | $E_\gamma = 26.35$ keV | |
| 10.81 | L_I | 33.22 | I | w | | | | | |
| 11.65 | L_{II} | 33.24 | I | vvw | | | | | |
| 15.64 | L_{III} | 33.25 | I | w broad | | | | Probably unresolved doublet | |
| 27.45 | M_I | 33.19 | I | ms | | | | | |
| 27.84 | M_{II} | 33.20 | I | m | | | | | |
| 28.77 | M_{III} | 33.20 | I | m | | | | | |
| 29.39 | M_{IV} | 33.16 | I | vvw | | | | | |
| 31.70 | N_I | 33.20 | I | m | | | | | |
| 31.87 | N_{II} | 33.19 | I | vw | | | | | |
| 32.11 | N_{III} | 33.19 | I | vw | | | | | |
| 32.89 | O | ~33.2 | I | w broad | | | | | |
| 33.18 | P | ~33.3 | I | vvw? | | | | $E_\gamma = 33.20$ keV | |
| 21.05 | L_I | 43.46 | I | m | | | | Composite with 26.4 M_{II} line | |
| 21.87 | L_{II} | 43.46 | I | m | | | | | |
| 25.85 | L_{III} | 43.46 | I | m | | | | | |
| ... | M_I | ... | I | ... | | | | Obscured by 59.6 L_{II} line | |
| ... | M_{II} | ... | I | ... | | | | Obscured by 59.6 L_{II} line | |
| 39.00 | M_{III} | 43.43 | I | wm | | | | N and O lines unresolved from 64.9 L_I and L_{II} lines | |
| ... | N | ... | I | ... | | | | | |
| ... | O | ... | I | ... | | | | | |
| 37.16 | L_I | 59.57 | I | vvs | | 695 | 2060 | $E_\gamma = 43.46$ keV | |
| 37.98 | L_{II} | 59.57 | I | vvs | | 1370 | | | |
| 41.96 | L_{III} | 59.57 | I | vs | | 435 | 315 | | |
| 53.83 | M_I | 59.57 | I | vs | | ~250(est.) | | | |
| 54.21 | M_{II} | 59.57 | I | vs | | 520 | | | |
| 55.14 | M_{III} | 59.57 | I | s | | 135 | 660 | | |
| 55.70 | M_{IV} | 59.55 | I | w | | | | | |
| 55.89 | M_V | 59.55 | I | w | | | | | |
| 58.07 | N_I | 59.57 | I | ms | | ~75(est.) | 144 | | |
| 58.23 | N_{II} | 59.55 | I | ms | | ~150(est.) | | | |
| 58.47 | N_{III} | 59.55 | I | w | | ~50(est.) | | | |
| 58.80 | N_{IV+V} | 59.57 | I | w | | | | | |
| 59.30 | O | ~59.6 | I | m broad | | | | | |
| 59.56 | P | ~59.6 | I | w | | | | | |
| 42.41 | L_I | 64.82 | I | w broad | | ~25(est.) | | $E_\gamma = 59.57$ keV | |
| 43.17 | L_{II} | 64.76 | I | w broad | | ~25(est.) | | L_I and L_{II} lines contain N and O lines of 43.5 γ , but intensity is mostly due to 64.8 γ | |
| 47.25 | L_{III} | 64.86 | I | w | | 25(est.) | | Obscured by O and P lines of 59.6 γ | |
| ... | M_I | ... | I | ... | | | | | |
| ... | M_{II} | ... | I | ... | | | | | |
| 60.46 | M_{III} | 64.89 | I | vw | | | | | |
| 63.72 | N | ~65.0 | I | vvw? | | | | $E_\gamma = 64.8$ keV | |
| 91.51 | L_I | 113.92 | II | | | | | | |
| 92.36 | L_{II} | 113.95 | II | | | | | | |
| 96.29 | L_{III} | 113.90 | II | | | | | | |
| 45.97 | K | 164.6 | I | wm | | 40-50(est.) | >5 <16 | $E_\gamma = 113.9$ keV | |
| 142.2 | L_I | 164.6 | II | | wm | 15.5 | 130 | | |
| 143.0 | L_{II} | 164.6 | II | | s | 140 | | | |
| 146.9 | L_{III} | 164.5 | II | | ms | 69 | 39 | | |
| 159.2 | M_{II} | 164.6 | II | | wm | 34 | 33 | | |
| 160.1 | M_{III} | 164.5 | II | | wm | 21 | | | |
| 163.3 | N_{II+III} | 164.4 | II | | w broad | | | | |
| 164.4 | O | ~164.7 | II | | vw broad | | | | |
| 89.3 | K | 207.9 | II | | vvvs | 4600 | 6790 | $E_\gamma = 164.6$ keV | |
| 185.6 | L_I | 208.0 | II | | vvs | 1000 | 1130 | Permanent-magnet and double-focusing intensities normalized here. | |
| 186.4 | L_{II} | 208.0 | II | | m | 130 | | | |
| 190.2 | L_{III} | 207.8 | II | | vw | 8.1 | 6.0 | | |

TABLE I.—Continued.

| Electron energy (keV) ^b | Shell ^c | Transition energy (keV) | Instrument ^d quoted | Visual intensity estimate ^a | | | Intensity (densitometer) | Intensity (double-focusing) | Remarks |
|------------------------------------|------------------------|-------------------------|--------------------------------|--|-------------------|--------------------|--------------------------|-----------------------------|---------------------------------------|
| | | | | PM I (plate 264) | PM II (plate 272) | PM III (plate 279) | | | |
| 202.1 | <i>M_I</i> | 207.8 | II | | s | | 225 | 193 | |
| 202.6 | <i>M_{II}</i> | 208.0 | II | | vw | | | | |
| 203.3 | <i>M_{III}</i> | 207.7 | II | | vvvw? | | | | |
| 206.4 | <i>N_I</i> | 207.9 | II | | m | | 75 | 62 | |
| 207.7 | <i>O</i> | 208.0 | II | | m | | | | |
| 115.6 | <i>K</i> | 234.2 | II | | w | | 12 | | <i>E_γ</i> = 207.9 keV |
| 211.8 | <i>L_I</i> | 234.2 | II | | vw | | | | |
| 148.8 | <i>K</i> | 267.4 | II | | m | | 50 | 25 | <i>E_γ</i> = 234.2 keV |
| 245.1 | <i>L_I</i> | 267.5 | II, III | | vw | s | 17 | } 11 | |
| 245.7 | <i>L_{II}</i> | 267.3 | III | | | vw | | | |
| ... | <i>L_{III}</i> | ... | III | | | | | | <i>L_{III}</i> line masked by |
| 261.8 | <i>M_I</i> | 267.5 | III | | | m | 6.5 | 7.0 | 368.5 _γ <i>K</i> line |
| 266.0 | <i>N</i> | 267.5 | III | | | w | | | |
| 267.3 | <i>O</i> | 267.6 | III | | | vw | | | <i>E_γ</i> = 267.5 keV |
| 213.5 | <i>K</i> | 332.2 | II, III | | vw | ms | 4.9 | | |
| 309.9 | <i>L_I</i> | 332.3 | III | | | w(?) | | | |
| 310.7 | <i>L_{II}</i> | 332.3 | III | | | m | 3.2 | | |
| 314.8 | <i>L_{III}</i> | 332.4 | III | | | w | | | |
| 327.0 | <i>M_{II}</i> | 332.4 | III | | | w | | | |
| 330.8 | <i>N_{II}</i> | 332.1 | III | | | vw | | | <i>E_γ</i> = 332.3 keV |
| 216.7 | <i>K</i> | 335.3 | III | | | ms | 3.7 | | |
| 312.8 | <i>L_I</i> | 335.2 | III | | | w | | | |
| 329.2 | <i>M_I</i> | 334.9 | III | | | vw? | | | <i>E_γ</i> = 335.3 keV |
| 249.9 | <i>K</i> | 368.5 | III | | | w | 2.2 | | |
| 346.3 | <i>L_I</i> | 368.7 | III | | | vw | | | <i>E_γ</i> = 368.5 keV |
| 252.3 | <i>K</i> | 370.9 | III | | | m | 4.1 | | |
| 348.5 | <i>L_I</i> | 370.9 | III | | | w | | | <i>E_γ</i> = 370.9 keV |

^a Unassigned lines

| <i>E</i> (keV) | Int. | Instr. |
|----------------|---------|--------|
| 9.99 | vw | I |
| 22.67 | w broad | I |
| 139.6 | vw | II |
| 200.5 | vw | III |
| 228.5 | vw | III |

^b Electron energies in italics are those lines used as internal standards. The corresponding gamma-ray energies were measured by P. P. Day, Phys. Rev. **97**, 689 (1955).^c Electron binding energies have been taken from the compilation of Hill, Church, and Mihelich, Rev. Sci. Instr. **23**, 523 (1952).^d PM I = 50 gauss, PM II = 100 gauss, PM III = 215 gauss.^e s = strong, m = moderate, w = weak, v = very ... These estimates are relative only for measurements made in the same spectrograph.

6*M* HCl containing a small amount of nitric acid. A separation of uranium from neptunium was then effected by passing the solution through a Dowex A-1 anion-exchange column; under these conditions neptunium is adsorbed by the resin while uranium is not. Further purification and concentration of the uranium was done by extraction into di-ethyl ether from ammonium nitrate solution followed by back extraction into water. This solution was evaporated to dryness, taken up in 0.5 ml of slightly-acidified ammonium oxalate solution (40 g/l), and then the uranium was electrodeposited upon a 10-mil platinum wire which served as the beta-spectrograph source. Sources for the other measurements were prepared similarly.

The decay of part of one of these sources was followed with a Geiger-Müller counter for six half-lives and showed no deviation from the accepted 6.8-day half-life.¹⁹

ELECTRON SPECTROSCOPY

Measurements were made with the following instruments:

¹⁹ J. R. Huizenga and K. F. Flynn (unpublished results, 1953).

1. The Berkeley permanent-magnet electron spectrographs were used to study the conversion-electron spectrum of U²³⁷. These are 180° photographic-recording instruments which operate at a momentum resolution of ~0.1%. Three spectrographs were used, of nominal field strengths 50, 100, and 215 gauss. The electrons are recorded on 25μ Eastman No-Screen X-Ray emulsion on glass backing of dimensions 3/4×15×0.04 inch. Their calibration and operation have been described by Smith and Hollander.²⁰

2. The Berkeley double-focusing spectrometer was used primarily to study the beta continuum and to provide additional information about the conversion-electron spectrum, especially with regard to intensities of conversion lines. This instrument, originally built and described by O'Kelley,²¹ has recently been improved by the installation of new acceptance and Geiger counter slits so that it presently operates at ~0.3% resolution and ~0.1% transmission. These modifications were carried out largely by Dr. S. Thulin.

²⁰ W. G. Smith and J. M. Hollander, Phys. Rev. **101**, 746 (1956)

²¹ G. D. O'Kelley, University of California Radiation Laboratory Report UCRL-1243, June, 1951 (unpublished).

TABLE II. Transition energies (keV) reported from recent electron-spectroscopic studies of U^{237} .

| Present work | Baranov and Shlyagin ^a | Bunker, Mize, and Starner ^b |
|--------------|-----------------------------------|--|
| 26.35 | 26.4 | 26.3 |
| 33.20 | 33.3 | 33.2 |
| 43.46 | 43.5 | 43.4 |
| 59.57 | 59.7 | 59.6 |
| 64.8 | ... | ... |
| ... | 69(?) | ... |
| ... | ~101(?) | ... |
| 113.9 | ... | ... |
| ... | ~124(?) | ... |
| 164.6 | 165.4 | 164.3 |
| ... | ~193(?) | ... |
| 207.9 | 208.2 | 207.7 |
| 234.2 | ... | 234.1 |
| 267.5 | 267.5 | 267.3 |
| 332.3 | 331.5 | 332.1 |
| 335.3 | ... | 335.2 |
| 368.5 | ... | 368.4 |
| 370.9 | 369.5 | 370.7 |
| ... | ~436 | ... |

^a See reference 15.^b See reference 18.

In Table I we have summarized the conversion-electron data. Except for the column labeled *Intensity* (*double-focusing*), all the data presented in this table were obtained with the permanent-magnet spectrographs. The absolute error of the energy values is estimated to be less than 0.2%, and for those electron lines in the vicinity of the internal standard lines the error should be less than 0.1%.

TABLE III. Comparison of some relative intensity measurements in decay of U^{237} normalized to $L_I + L_{II}(208) = 1130$.

| Conversion lines E_γ | Shell | Wagner <i>et al.</i> | Baranov and Shlyagin | Double-focusing spectrometer | Permanent-magnet spectrograph | | | |
|-----------------------------|-----------|----------------------|----------------------|------------------------------|-------------------------------|-----|-----|-----|
| 59 | L_I | }1540 | }2130 | }2060 | 695 | | | |
| | L_{II} | | | | 1370 | | | |
| | L_{III} | | | | 352 | 315 | 435 | |
| | M | | | | 514 | 652 | 655 | |
| | N | ... | 276 | 144 | ... | | | |
| 164 | K | ... | ... | 5 to 16 | 40-50 | | | |
| | L_I | }257 | }138 | }130 | 15.5 | | | |
| | L_{II} | | | | 140 | | | |
| | L_{III} | | | | ... | 83 | 39 | 69 |
| | M | | | | 31 | 44 | 33 | 56 |
| 208 | K | 5449 | 4400 | 6790 | 4600 | | | |
| | L_I | }1130 | }1130 | }1130 | 1000 | | | |
| | L_{II} | | | | 130 | | | |
| | L_{III} | | | | ... | ... | 6.0 | 8.1 |
| | M | | | | 308 | 251 | 193 | 225 |
| | N | ... | 95 | 62 | 75 | | | |
| 267 | K | ... | ... | 25 | 50 | | | |
| | L_I | 23 | ... | 11 | 17 | | | |
| | M_I | 7 | ... | 7.0 | 6.5 | | | |
| 368 | K | }2 | ... | 2.0 | 2.2 | | | |
| 370 | K | | ... | 2.0 | 4.1 | | | |
| β Groups | | | | | | | | |
| 248 | | 10 280 | 12 550 | 15 000 | ... | | | |
| 81 | | ≤ 3055 | 4400 | < 1200 | ... | | | |
| (450) | | < 10 | none | < 210 | ... | | | |

Relative intensities of lines recorded in the permanent-magnet spectrographs were measured by the densitometer method of Mladjenovic and Slätis,²² in which the intensity is given by $I = A\rho/\eta$, where A \equiv height of line \times half-width, ρ \equiv radius of curvature, and η \equiv energy efficiency of emulsion.

Conversion-electron intensities from spectra obtained with the double-focusing spectrometer were measured by integrating the areas under conversion-line peaks with a planimeter and dividing by the $H\rho$ value of the line. Although the large number of conversion lines obscures considerable portions of the beta continuum, a reconstruction of the spectrum made from the Fermi plot allowed the contribution of the continuum to be subtracted out from the total observed spectrum to obtain the contribution of the conversion lines.

Table II gives a comparison of some of the gamma transition energies reported in recent electron-spectroscopic studies of U^{237} .

Table III gives a comparison of some of the relative electron intensities observed in this work with those reported by Wagner *et al.*¹⁴ and by Baranov and

TABLE IV. Auger lines from U^{237} decay.

| Transition | $E_{exp.}$ (keV) | $E_{theor.}^a$ | Visual intensity estimate |
|-----------------------|------------------|----------------|--------------------------------------|
| $K - L_I L_I$ | 73.41 | 73.41 | m |
| $K - L_I L_{II}$ | 74.25 | 74.24 | ms |
| $K - L_I L_{III}$ | 78.21 | 78.26 | wm |
| $K - L_{III} L_{II}$ | 78.95 | 79.04 | m |
| $K - L_{III} L_{III}$ | 82.99 | 83.06 | wm |
| $K - L_I M_I$ | ... | 90.5 | ... masked by 207.9- γ K line |

^a I. Bergström and R. D. Hill, *Arkiv Fysik* **8**, 2, 21 (1954).

Shlyagin.¹⁵ The intensities of all determinations are normalized to 1130 total intensity for L_I plus L_{II} lines of the 208-keV transition. The agreement among the various determinations is only moderate and probably gives a realistic measure of the absolute accuracy of the intensities. For the later sections of this paper where electron intensities are needed we use an average of our double-focusing and 180° permanent-magnet intensities, with subshell ratios as measured in the permanent-magnet spectrographs.

Out of a total of 93 lines observed in the 180° spectrographs, all but five have been assigned to 14 gamma rays and the K -Auger lines. The Auger line energies and intensities are given in Table IV.

A Fermi plot of the beta continuum, obtained with the double-focusing spectrometer, yielded an end point of 248 keV, in good agreement with the result of Wagner *et al.*,¹⁴ 245 keV, and that of Baranov and Shlyagin,¹⁵ 249 keV.

A search was made for a spectrum of about 450 keV, corresponding to decay to the 60-keV level; such a group was not found and an upper limit on its abun-

²² M. Mladjenovic and H. Slätis, *Arkiv Fysik* **8**, 65 (1954).

dance per disintegration was set at 1%. Wagner *et al.*¹⁴ set a much lower limit, <0.1%, in searching for a 511-keV beta group, a limit which should apply to the possible 450-keV group as well.

Wagner *et al.*¹⁴ cite evidence for a lower energy component in the beta spectrum with abundance between 5 and 20%. Baranov and Shlyagin¹⁵ interpret an excess of electrons in the low-energy portion of the Fermi plot as due to an 86-keV spectrum occurring in 26% abundance. The latter results are inconsistent with the present observations, which indicate that the low-energy beta components have a total intensity of around 4%. Our figure is based not a subtracted beta spectrum but rather on a total intensity for all gamma rays which originate from levels higher than the heavily populated 267.5-keV level.

GAMMA-RAY SPECTROSCOPY

Gamma-ray spectroscopic measurements were carried out with a scintillation spectrometer employing a 1½ in. diameter by 1 in. thick sodium iodide (thallium-activated) crystal with a Dumont 6292 photomultiplier tube, an Oak Ridge-type double-differentiating linear amplifier, and a Penco 100-channel pulse-height analyzer. The Penco analyzer utilizes pulse-height-to-time conversion and a magnetic-core memory-storage unit; the analyzer channels have extremely uniform window widths. This equipment yielded 8.5% energy resolution on the 662-keV gamma ray of Cs^{137} .

Gamma rays of energies 60, 102 (*K* x-rays), 163, 208, ~266, and 332 keV were clearly present, and by careful study of the spectrum transmitted through lead absorbers of thicknesses up to 8 g/cm² the existence of radiation of about 365-keV energy was shown. Of these, the 266- and 365-keV photons have not been reported previously from scintillation spectroscopy.

Table V lists the measured photon energies, the more precise corresponding transition energies as determined from the electron spectra, and the photon intensities of Wagner *et al.*¹⁴ and of the present work. All photon intensities have been normalized to an absolute intensity of 38 photons per 100 disintegrations for the 60-keV transition. This figure is based upon the absolute abundance of this gamma ray in Am^{241} decay, 37 photons per 100 disintegrations, measured by Magnusson.²³ There are two corrections applying to the normalization for U^{237} . One of these results from the fact that approximately 4% of the U^{237} disintegrations (our result) bypass the 59.6-keV state, whereas virtually all (99.3%) of the Am^{241} disintegrations pass through it. The other correction arises because about 6% of the 60-keV photon peak in U^{237} decay consists of another gamma ray (64.9 keV).

COINCIDENCE STUDIES

Gamma-gamma coincidence measurements were carried out by Dr. F. Asaro with apparatus employing two

²³ L. B. Magnusson (private communication, March, 1956).

TABLE V. Electromagnetic radiation of U^{237} from scintillation spectra.

| Measured photon energy (keV) | Transition energies from electron spectra | Photons/100 beta disintegrations ^b | |
|------------------------------|---|---|-------------------|
| | | Wagner <i>et al.</i> | This work |
| 60 | 59.57 } (64.8) } | 38 | 38 ^c |
| 102 ^a | ... | 54 | 55 |
| 163 | 164.6 | ... | 3.6 |
| 208 | 207.9 | 22 | 24 |
| ~266 | 267.4 | ... | 0.86 |
| 332 | 332.3 } 335.3 } | 2.7 | 1.57 ^d |
| ~365 | 368.5 } 370.9 } | ... | ~0.10 |

^a *K* x-rays.

^b Relative intensities normalized to 0.38- {59.6-keV + 64.8-keV} photons per disintegration.

^c From coincidence work, discussed later, it can be determined that the intensity ratio of 59.6- to 64.8-keV photons is 16:1.

^d From coincidence work, discussed later, it can be determined that the intensity ratio of 332.3- to 335.3-keV photons is 7.3:1.

1 in. thick by 1½ in. diameter sodium iodide crystal detectors with Dumont 6292 photomultiplier tubes. Pulse-amplitude discrimination was made in the "gate" channel by a single-channel pulse-height analyzer, and the coincidence spectrum from the other channel was displayed on a 50-channel pulse-height analyzer. The coincidence apparatus was operated at a resolving time (2τ) of 3×10^{-6} sec.

The principal results are summarized in Table VI. These coincidence results are fully consistent with and have helped to establish the decay scheme of Fig. 1. In attempting to interpret the changes in relative gamma intensities between coincidence and "singles" spectra, one should note that the observed photon peaks in two cases are composed of two unresolved gamma rays. The fact that the coincidence measurements with photon gate labeled 60 keV include both the 59.6-keV and 64.9-keV *E1* gammas explains the presence of weak (60-keV) coincidences with the 267.4-keV gamma. In the "singles" spectrum the peak at ~332 keV is composed of 332.3- and 335.3-keV photons. The coincidence measurements with *L* x-ray gating can be interpreted as showing that the more abundant of these gamma rays (the 332.3-keV) proceeds directly to ground; the fact

TABLE VI. Results of gamma-gamma coincidence measurements.

| Photon "gate" (keV) | Coincident gamma rays ^a (keV) |
|---------------------|--|
| <i>L</i> x-rays | <i>K</i> x-ray 163 208 332 ^b |
| 60 | <i>K</i> x-ray 163 208 266 ^b |

^a The coincident gamma spectrum was not examined at energies much below the *K* x-ray energy. One would, of course, expect that there would be coincidences between *L* x-rays and 60-keV gammas, but these were not looked for.

^b Intensity of each of these gammas relative to the 208 in the coincidence spectrum is much less than in the "singles" spectrum.

332.3-keV transition proceeds directly to the ground state, an interpretation which is supported by the additional observation of coincidences between photons of ~ 60 keV and ~ 270 keV. The sum of the measured energies, $64.9 \text{ keV} + 267.5 \text{ keV} = 332.4 \text{ keV}$, is also in good agreement with the measured energy of the crossover gamma ray, 332.3 keV.

Wagner *et al.*,¹⁴ from conversion-line data, proposed gamma transitions of 334 and 370 keV in U²³⁷ decay. These transitions they postulated as arising from a level at 431 keV. Baranov and Shlyagin¹⁵ measured these transitions as 330 and 369 keV, and also placed their origin at a 433-keV level. In the present work, the energies of these transitions have been measured as 335.3 and 368.5 keV; and the 33.2-keV difference between these values coincides with the energy of the 33.20-keV transition and strongly suggests that the 33.2 and 335.3 are cascade gamma rays, with the 368.5 as crossover. A new level would thus be defined as 368.5 keV. Some independent evidence for this state is provided by the gamma-gamma coincidence result that although most of the ~ 335 -keV radiation appears to go to ground (the 332.3-keV transition as discussed above), a small fraction of these photons appears to be in coincidence with *L* x-rays but not with 60-keV radiation. Hence the 335.3-keV transition probably goes to the 33.2-keV level. With this interpretation we may make use of the relative-intensity information from coincidence work to calculate that the 335-keV peak in the "singles" gamma spectrum is composed of unresolved 332.3-keV and 335.3-keV peaks having relative intensities 88% and 12%, respectively. No coincidences could be found with 370-keV photons, which is additional evidence that the 368.5-keV transition goes to ground.

We observe also a 370.9-keV transition. Because of the absence of coincidences of 370-keV photons with any other radiation, just discussed, it is also assumed that the 370.9-keV photon goes to ground, thus defining a new level at that energy.

Wagner *et al.*¹⁴ report coincidences between the 208-keV photon and a photon of 165 keV, with the interpretation that there is a level at 430 keV. The present results do not confirm this evidence, for two reasons. First, we see in the electron spectrum only one gamma ray of ~ 165 keV, the 162.5-keV transition, which has been unambiguously placed in the scheme *in parallel* with the 208-keV transition. Second, as mentioned in the earlier section on coincidence measurements, Asaro could find no true coincidences between 208- and 165-keV gammas and was able to set a low limit on their possible existence.

The only gamma ray which we have not placed in the above scheme is a very weak transition of 113.9 keV, observed only in the electron spectrum.

Although we do not have the detailed evidence by which Bunker *et al.*¹⁸ arrive at a level scheme, we wish to mention that their postulated level system differs from ours only in that they included a level at 335.9

TABLE VII. *L* and *M* subshell conversion ratios of 60-keV gamma ray in Np²³⁷.

| Subshell | Relative coefficients | Source | Reference |
|---|-----------------------|--|-----------------------------------|
| <i>L</i> _I : <i>L</i> _{II} : <i>L</i> _{III} | 1.6:3.2:1.0 | U ²³⁷ | This work |
| | 1.5:3.3:1.0 | Am ²⁴¹ | HSR ^a |
| | 2.2:4.7:1.0 | Am ²⁴¹ | Baranov and Shlyagin ^b |
| | 2.4:4.7:1.0 | Am ²⁴¹ | Canavan ^c |
| | 1.0:1.0:1.0 | Theoretical (screened relativistic point nucleus) | Rose ^d |
| <i>M</i> _I : <i>M</i> _{II} : <i>M</i> _{III} : <i>M</i> _{IV+V} | 1.7:3.6:1.0:0.1 | Am ²⁴¹ | HSR ^a |
| | 1.1:0.9:1.0:0.4 | Theoretical (unscreened, relativistic point nucleus) | Rose ^e |

- ^a See reference 1.
^b See reference 15.
^c F. L. Canavan (unpublished results, 1956).
^d See reference 26.
^e M. E. Rose (privately circulated tables).

keV, whereas we assign the transition of that energy to proceed from the 368.5-keV level to the 33.2-keV level.

CONVERSION COEFFICIENTS, MULTIPOLARITIES, AND SPINS

Ground state.—The spin of Np²³⁷ has been measured as 5/2 by Tomkins.²⁴ We shall assume even parity for the ground state as postulated by HSR,¹ but in this section only the relative parities of the states will actually be considered.

33.2-keV state.—The multipolarity of the 33.2-keV gamma ray has been determined as mixed *M*₁–*E*₂ by measurement of the *L*- and *M*-subshell conversion ratios from Am²⁴¹ decay¹; the mixing ratio was reported as *M*₁/*E*₂ ~ 50 . From the large corrections to the magnetic-dipole *K*-conversion coefficients occasioned by the finite nuclear-size effects treated by Sliv,²⁵ it seems likely that the *L*_I-subshell conversion coefficients will be similarly affected; the *M*₁/*E*₂ photon ratio should accordingly be increased to ~ 90 . The present data on U²³⁷ are in agreement with but add nothing to our present knowledge of this transition. The spin of this state is 7/2+.

59.6-keV state.—The 59.6-keV electric dipole transition from this 5/2 state to ground has been studied extensively from Am²⁴¹ alpha decay; a discussion of previous work is given in HSR.¹ The present studies of U²³⁷ confirm the fact, pointed out by HSR, that the relative *L*- and *M*-subshell internal conversion coefficients are in marked disagreement with the theoretical values of Rose²⁶ for an *E*₁ transition of 60 keV in *Z* = 93. Table VII summarizes the experimental and theoretical values. It is noted that the *M*_I:*M*_{II}:*M*_{III} ratios follow

²⁴ F. S. Tomkins, Phys. Rev. **73**, 1214 (1948).

²⁵ L. A. Sliv, privately circulated tables of relativistic, screened *K*-conversion coefficients for nuclei of finite size.

²⁶ M. E. Rose, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), App. IV, p. 905, and privately circulated tables.

closely the L ratios, and are correspondingly anomalous; M_{IV+V} conversion is weaker by a factor of four relative to M_{III} than the theoretical value.

The 26.4-keV transition is known also to be electric dipole from its position in the level scheme.^{1,2,7} Because of the anomalous L -subshell ratios of the 59.6-keV transition, it is of interest to examine the subshell ratios of this transition. Our M -subshell ratios are uncertain because of the accidental coincidence in energy of the M_{II} and M_{III} lines of the 26.4-keV gamma with the L_I and L_{II} lines of the 43.5-keV gamma. However, the N_I , N_{II} , and N_{III} lines are completely resolved; their intensity ratios are estimated visually to be of the order $N_I:N_{II}:N_{III}\sim 1.7:3.3:1.0$. The theoretical L -subshell ratios of Rose²⁶ for this gamma ray are 0.56:1.1:1.0, and the theoretical M -subshell ratios²⁷ are 0.6:0.8:1.0:0.47:0.55. Thus, also in this case the subshell conversion ratios are somewhat at variance with the theoretical values.

For calculation of the relative intensities of radiation depopulating the 60-keV state, we choose to use intensity data of other workers. Magnusson²³ has determined by scintillation spectroscopy that there are 0.37 photons of 60 keV per Am²⁴¹ alpha disintegration. From alpha-ray spectroscopy it is known² that 99.3% of the alpha disintegrations cascade through the 60-keV state. From the electron-spectroscopic work of Baranov and Shlyagin¹⁵ on Am²⁴¹ we have taken the ratio of total conversion lines of the 60-keV to the 33.2-keV transition. The 33.2-keV transition is almost totally converted.

From the above information we calculate that the depopulation of the 60-keV state occurs 84% by the 60-keV transition and 16% by the 26-keV, 33-keV cascade. One can also calculate a total conversion coefficient for the 60-keV transition of 1.3. Using Magnusson's figure of 0.028 for the number of 26-keV photons per alpha a total conversion coefficient of about 4.7 is calculated for that transition.

103.0-keV state.—The 43.4-keV gamma ray, which de-excites this state, has been determined from Am²⁴¹ decay¹ to be an $M1$ - $E2$ mixture, with $M1/E2\sim 6$. (We estimate that the $M1/E2$ ratio will be increased to ~ 11 when the effects of finite nuclear size correction are taken into account.) This gamma ray was also seen in the present study of U²³⁷, but no new multipolarity information has been obtained. The spin of this state is thought¹ to be $7/2^-$.

267.5-keV state.—Definite multipolarity assignments can be made for the gamma rays which de-excite this state. The 207.9-keV transition is shown definitely to be magnetic dipole from the following information:

The L -subshell ratios, $L_I:L_{II}:L_{III}=7.7:1.0:\sim 0.06$, are consistent only with an interpretation as predominantly $M1$. For an $M1$ transition of this energy in $Z=93$ the theoretical ratios of Rose²⁶ are $L_I:L_{II}:L_{III}=10.4:1.0:0.03$. A reduction of 26% in the $L_I:L_{II}$

ratio from Rose's theoretical value is indicated by our results. Perhaps the agreement between theory and experiment will improve with the use of L -shell coefficients which are corrected for the effects of finite nuclear size, since these corrections would be expected theoretically to lower principally the L_I conversion coefficient. However, Sokolowski *et al.*²⁸ have found, in contrast to this expectation, that the L_I/L_{II} ratio of the 40-keV $M1$ transition in ThB is experimentally the same as that calculated from Rose's point-nucleus conversion coefficients. It should also be noted that this 208-keV $M1$ transition from U²³⁷ decay is highly retarded (factor of 10^4), and conceivably the model-dependent effects on conversion coefficients discussed by Church and Weneser²⁹ might not be negligible.

The very low intensity of the L_{III} line of this transition shows that there is very little $E2$ admixture. From the experimental value of the absolute L_{III} conversion coefficient from Table VIII and Rose's theoretical values we estimate the $E2/M1$ photon mixing ratio as 0.005 ± 0.005 .

The absolute K -conversion coefficient of the 207.9-keV gamma ray has been measured by Dr. D. Strominger from the relative K x-ray and 207.9-keV gamma-ray intensities in the U²³⁷ photon spectrum. The calculation is facilitated by the fact that the K vacancies are produced predominantly by conversion of this gamma. Small corrections have been applied for K vacancies arising from other transitions and for the K -fluorescence yield. The experimental value, $e_K/\gamma=2.3$, is in good agreement with the theoretical $M1$ K -conversion coefficient of Sliv,²⁵ $\beta_1=2.4$.

With the multipolarity of the 207.9-keV transition definitely assigned, it becomes possible to use our conversion-coefficient information and the electron and photon intensities to calculate conversion coefficients of the other gamma rays. There are several alternative ways to do this. One might use the measured (or theoretical) K -conversion coefficient of the 207.9-keV gamma as a means to normalize electron intensities relative to photon intensities and thereby obtain conversion coefficients of the other gamma rays. In the present case, this method does not seem attractive because there exists a wide discrepancy among the various determinations of the K -line intensity of this gamma ray (see Table III) relative to all other lines, and hence any average we might select would be arbitrary. Alternatively, one might utilize our measured L_I -line intensity of this gamma (which we feel is more reliable experimentally), and normalize this to the Rose theoretical L_I -conversion coefficient; this method is not satisfactory either, because of the magnitude of the finite-size correction expected for the magnetic-dipole L_I coefficients. A third method, which we have chosen to use, is to assume that Rose's theoretical K/L_I ratio

²⁸ Sokolowski, Edvarson, and Siegbahn, *Nuclear Phys.* **1**, 160 (1956).

²⁹ E. L. Church and J. Weneser, *Phys. Rev.* **104**, 1382 (1956).

²⁷ M. E. Rose (privately circulated tables).

TABLE VIII. Percentage intensities and experimental conversion coefficients.

| Transition energy (kev) | K | Conversion electron intensities | | | | Total | Photon intensity ^d | Experimental conversion coefficients | | | Multi-polarity | Total estimated transition intensity | |
|-------------------------|-------|---------------------------------|----------|-----------|------------------|-------|-------------------------------|--------------------------------------|-------|----------|----------------|--------------------------------------|-----------|
| | | L_I | L_{II} | L_{III} | M | | | K | L_I | L_{II} | | | L_{III} |
| 26.35 | | | | | a | | | | | | $E1$ | 15 | |
| 33.20 | | a | a | a | a | | | | | | $M1+E2$ | 15 | |
| 43.46 | | a | a | a | a | | | | | | $M1+E2$ | 7 | |
| 59.57 | | 7.5 | 14.9 | 4.7 | 8.5 | 38.3 | 36 | | 0.21 | 0.41 | 0.13 | $E1$ | 81 |
| 64.8 | | ~0.27 | ~0.27 | ~0.27 | a | 1.1 | ~2.3 ^e | | ~0.12 | ~0.12 | ~0.12 | $E1$ | 3 |
| 113.9 | | a | a | a | | | | | | | | | |
| 164.6 | ~0.48 | 0.16 | 1.27 | 0.58 | 0.48 | 3.3 | 3.6 | ~0.13 | 0.04 | 0.35 | 0.16 | $E2$ | 7 |
| 207.9 | 50-74 | 10.8 ^b | 1.41 | 0.076 | 2.2 ^c | ... | 24 | 2.3 ^e | a | 0.059 | 0.003 | $M1$ | 89 |
| 234.2 | 0.13 | a | | | | | | | | | | $M2$ | ~0.2 |
| 267.5 | 0.54 | 0.16 | a | | 0.08 | 0.80 | 0.86 | 0.63 | 0.19 | | | $E1+M2$ | 1.7 |
| 332.3 | 0.053 | a | 0.035 | a | a | ~0.12 | 1.4 ^f | 0.038 | | 0.025 | | $E2$ | 1.5 |
| 335.3 | 0.040 | a | | | a | ~0.05 | 0.19 ^f | 0.2 | | | | $M1+E2$ | ~0.2 |
| 368.5 | 0.024 | a | | | | ~0.03 | | >0.24 | | | | $M1$ | ~0.05 |
| 370.9 | 0.044 | a | | | | ~0.06 | 0.10 | >0.44 | | | | $M1$ | ~0.10 |

^a Electron line observed but no reliable numerical intensity available from the present work. See Table I for visual estimate.

^b Electron intensities normalized to L_I -conversion coefficient of 0.45, obtained from experimental K -conversion coefficient (2.3) and theoretical K/L_I ratio (5.16) as explained in the text.

^c M -subshell ratios may be found from Table I.

^d Normalized to 36 60-kev photons per 100 betas.

^e The intensity of 64.8-kev photons is based on coincidence intensities and is subject to perhaps $\pm 30\%$ uncertainty.

^f Total intensity of the unresolved 335-kev photon peak has been divided between 332.3- and 335.3-kev transitions in the ratio obtained from the coincidence measurements.

^g Experimental value, from scintillation spectrometry.

for an $M1$ transition will be unaltered by the finite nuclear-size correction and apply this ratio to normalize the L_I electron intensity, using our experimental K -conversion coefficient of 2.3 and the value (Table V) of 24 photons (208 kev) per 100 beta disintegrations. The beta-spectroscopic work of Sokolowski *et al.*²⁸ on ThB strongly supports the assumption that the K -to- L_I conversion ratio for a pure $M1$ transition in a heavy element is correctly given by Rose's point-nucleus calculations. Interpolating in energy and atomic number from Rose's tables one finds a theoretical K/L_I ratio here of 5.16.

Table VIII is a summary of electron and gamma-ray intensities; electron intensities are normalized to 10.8 L_I (208) electrons per 100 betas and gamma intensities are normalized to 36 (59.6-kev) photons per 100 betas. The reasons for the choices of normalization have already been discussed. The electron intensities in Table VIII in most cases are an average of our permanent-magnet spectrograph intensities and our double-focusing spectrometer intensities except that the subshell ratios determined on the former instruments are used to divide the average total intensity of L - and M -shell conversion electrons.

The electron intensities are divided by the corresponding photon intensities to give the absolute conversion coefficients listed in Table VIII. The figures for total transition intensities, given in the last column of Table VIII, are for the weaker transitions just the sums of electron and photon intensities from this table. For the 207.9-kev transition the intensity is based on the requirement of intensity balance to and from the 60-kev level. Information from studies on Am^{241} , previously discussed, was used to establish relative intensities of the 26-kev and 33-kev transitions.

Multipolarities of the other gamma rays de-exciting the 267.5-kev state can now be discussed. The 164.6-kev gamma seems unambiguously to be electric quadrupole. The experimental L -conversion ratios of this transition are $L_I:L_{II}:L_{III}=0.12:1.0:0.5$; these values agree closely with the Rose theoretical values²⁶ for an $E2$ transition, which are $L_I:L_{II}:L_{III}=0.11:1.0:\sim 0.6$. The agreement of the experimental K -conversion coefficient, 0.12-0.15, with the Sliv²⁵ theoretical value, 0.19, is only fair, but probably is within experimental uncertainty. The absolute values of experimental and theoretical L_I , L_{II} , and L_{III} coefficients for $E2$ fail to agree by about a factor of two (experimental values too low).

The 234.2-kev transition is very weak, and the photon could not be resolved in the scintillation spectrum. Hence, we have no absolute conversion-coefficient information about it. From the level scheme, however, it will be established that this transition must be an $M2$. The observation of only one L -subshell line, the L_I , is consistent with the $M2$ assignment.

From its position in the level scheme, the 267.5-kev transition must be $E1$, $M2$, etc., because of the parity change involved (see Fig. 1). The experimental K -conversion coefficient (0.63) is intermediate between the Sliv value for $E1$ (0.040) and for $M2$ (3.6), indicating a photon mixing ratio $M2/E1\approx 0.16$. In like manner, from a comparison of the experimental L_I coefficient (0.19) with the Rose values for $E1$ (0.006) and $M2$ (1.2), we obtain a mixing ratio $M2/E1=0.15$. These two experimental values of the mixing ratio are essentially equal. In view of the fact that anomalously large L -conversion coefficients for $E1$ transitions have been observed from Am^{241} decay² and Th^{231} decay,³⁰ our

³⁰ Asaro, Stephens, and Perlman (unpublished data, 1956).

apparent photon ratio $M2/E1 \sim 0.16$ may be too high. This matter will be further discussed in the later section on comparisons with theory.

The small L_I conversion coefficient of the 164.6-keV transition indicates very little, if any, $M1$ mixing in this $E2$ transition. If one is permitted the assumption that this transition is *pure E2*, then the respective $M1$ and $E1$ characters of the 207.9- and 267.5-keV transitions allow a unique assignment of spin and parity $3/2^-$ to be made to the 267.5-keV state. If, on the other hand, one must admit the possibility of a large retardation of a hypothetical $M1$ component of the 164.6-keV transition (so that only the $E2$ component is observed) then the 267.5-keV state can only be labeled $3/2$, $5/2$, or $7/2^-$. There is additional evidence, however, which rules out the last two alternatives. This is the fact that the $9/2^-$ state¹ at 158.5 keV is not populated in U^{237} decay; were the spin of the 267.5-keV state $5/2^-$ (or larger), there would be easily detectable $E2$ (or $M1$) radiation to the $9/2^-$ state. Thus, it seems safe to conclude that the spin of the 267.5-keV state is $3/2^-$.

332.3-keV state.—The next higher state is at 332.3 keV. De-excitation takes place by a 332.3-keV transition to ground and by a 64.8-keV transition to the $3/2^-$ level. On the basis of the 332.3-keV gamma's prominent L conversion in the L_{II} subshell and its experimental K -conversion coefficient we assign it as $E2$. The agreement of the experimental K -conversion coefficient ($\alpha_K \approx 0.038$) with Sliv's theoretical $E2$ ($\alpha_K = 0.059$) coefficient is not good, but the experimental value is perhaps uncertain to this extent. The experimental L_{II} -conversion coefficient ($\alpha_{L_{II}} \approx 0.025$) is also lower than Rose's theoretical value ($\alpha_{L_{II}} = 0.044$).

The 64.8-keV transition, decaying in parallel with the 332.3-keV transition, can only be an $E1$ transition on the basis of the absolute L -subshell conversion coefficients as well as the L -subshell ratios (see Table VIII). Rose's theoretical screened-conversion coefficients for a 64.8-keV $E1$ transition ($Z=93$) should be as follows: $\alpha_{L_I} = 0.11$, $\alpha_{L_{II}} = 0.10$, $\alpha_{L_{III}} = 0.092$. It is interesting that this $E1$ transition exhibits normal L -conversion coefficients, while the 59.6-keV $E1$ transition exhibits a normal L_{III} but abnormally large L_I - and L_{II} -conversion coefficients. We believe the explanation may lie in the importance of model-dependent matrix elements for s_3 and p_3 electrons in retarded $E1$ transitions, these matrix elements being related in nature to those discussed by Church and Weneser²⁹ for $M1$ transitions.

A few words are in order at this point regarding the coincidence determination of relative amounts of 59.6-keV and 64.8-keV photons in the unresolved 60-keV peak in the "singles" gamma spectrum. As mentioned in the earlier section on coincidence measurements, the 267-keV to 208-keV photon ratio drops to 6% of its singles spectrum value when coincidence gating on the 59.6-keV and 64.8-keV photons is employed. Inspection of the decay scheme of Fig. 1 shows that events gated by 64.8-keV photons should display the same 267-keV

to 208-keV intensity ratio as "singles," while events gated by 59.6-keV photons should display a 208-keV peak but no 267-keV peak at all. Furthermore, the decay scheme shows that the fraction of 208-keV photon coincidence events per 64.8-keV photon should be very nearly the same as the events per 59.6-keV. Therefore, it directly follows that 6% of the composite photon peak at ~ 60 keV is 64.8-keV photons and the remaining 94% is 59.6-keV. This result was used in calculating photon intensities of Table VIII and in determining the proper normalization for intensities of Table V.

The multipolarity assignments to the 332.3-keV and 64.8-keV transitions are consistent with the fact that the final states of the two transitions are of opposite parity. The parity of the 332.3-keV state is even.

A rigorous spin assignment cannot be made to the 332.3-keV state on the basis of the above assignments alone; the values $1/2$, $3/2$, or $5/2+$ are possible since the possibility of small $M1$ admixture in the 332.3-keV $E2$ transition cannot strictly be ruled out. But $3/2$ or $5/2+$ are unlikely choices because a state with either of these spins would be expected to undergo appreciable $E2$ branching³¹ to the 33.2-keV $7/2+$ rotational state; such branching has not been observed. We therefore feel that the most reasonable assignment for the 332.3-keV state is $1/2+$.

368.5-keV state.—One has next the state of 368.5 keV, depopulated to the ground and first rotational states by gamma rays of 368.5 keV and 335.3 keV, respectively. From Table VIII we have a K -conversion coefficient of the 335.3-keV transition of $\alpha_K \approx 0.2$. The theoretical conversion coefficients of Sliv are 0.059 for $E2$ and 0.6 for $M1$. The observation of prominent L_I conversion and absence of L_{III} is not inconsistent with the assignment of this transition as mixed $M1$ - $E2$, but the uncertainty in the experimental K -conversion coefficient leaves the mixing ratio indefinite.

K -conversion coefficients cannot be calculated in the case of the 368.5-keV and 370.9-keV transitions since they are unresolvable in the photon spectrum. Both transitions exhibit similar conversion patterns (K/L ratio and prominent L_I conversion) and are probably of the same multipole order. Again, the prominence of the L_I conversion indicates that the transitions are either $E1$ or $M1$. A choice can easily be made from the limits on the absolute K -conversion coefficients. In Table VIII lower limits are given which definitely rule out $E1$; hence $M1$ assignments for both transitions seem best. The observed photon-intensity sum is consistent with the $M1$ assignments within the rather large uncertainty of both the photon and electron intensities of these weak transitions.

With the presence of $M1$ radiation in both the 335.3- and 368.5-keV transitions, the spin and parity of the 368.5-keV level can be either $5/2$ or $7/2+$.

370.9-keV state.—The only transitions not accounted

³¹ Alaga, Alder, Bohr, and Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 29, No. 9 (1955).

for with the levels heretofore postulated are those of 370.9 keV (*M*1) and of 113.9 keV (*E*2). Neither of these transitions can be related to a level scheme by energy-sum relations. We choose to list the 113.9-keV transition as unassigned, but there is evidence from our coincidence work that neither the 368.5- nor the 370.9-keV transitions are in coincidence with *L* x-rays. Hence, these transitions presumably go directly to ground. The spin of the 370.9-keV level can be 3/2, 5/2, or 7/2+. It is tempting to postulate that the 1/2+ state at 332.3 keV is the fundamental state of a *K*=1/2 rotational band and that the 3/2+ state is found at 370.9 keV, while the 5/2+ member lies just below it at 368.5 keV. This conjecture will be discussed further in the section on comparisons with theory.

BETA GROUP *ft* VALUES

As mentioned in the section on the beta spectrum, the Fermi plot was essentially straight in the region where the continuum was measurable, namely, above 90 keV (the energy of the strong 207.9 *K* line). The end point for the dominant beta group is about 248 keV. Our Fermi plot is not resolvable into the weaker lower energy beta branches, although it is consistent with their presence. Hence, we have calculated their intensities from the gamma-transition intensities and the proposed decay scheme. The beta-decay information is summarized in Table IX.

The failure to observe a beta group in the 410–510 keV energy range (spins of final states 5/2 or greater) strongly suggests that the spin of U²³⁷ is low, probably 1/2.

CORRELATIONS WITH THEORY

The most applicable nuclear model for Np²³⁷ is that of Bohr and Mottelson³² for nuclei with large spheroidal deformation. This section will discuss first the more general correlations with the model, considering rotational-band spacings and relative-intensity relationships involving the *K*-quantum numbers. Later the more special considerations depending on details of the state of intrinsic motion of the odd nucleon will be introduced. These latter considerations involve (1) association of observed band with proton eigenstates calculated by Nilsson,³³ (2) consideration of gamma-transition rates in terms of asymptotic quantum-number selection rules, and (3) consideration of beta *ft* values.

General Correlations

This study of U²³⁷ decay adds nothing to the knowledge of level spacings in the rotational bands based on ground and first excited states. One may refer to earlier publications^{1,2,6} for such data. We here hypothesize that the levels at 332.3, 368.5, and 370.9 keV, assigned

TABLE IX. Observed and postulated beta transitions of U²³⁷.

| Energy (keV) | Decay percent | Log <i>ft</i> | Final state, spin and parity |
|--------------|-------------------|---------------|------------------------------|
| 248 | 95 | 6.2 | 3/2- |
| (183) | ≤5 | ≥7.2 | 1/2+ |
| (144) | <5 and >0.1 | >6.9 and <8.5 | 3/2+(?) |
| (410) | <1 ^a | >8.9 | 5/2, (7/2)- |
| (510) | <0.1 ^b | >10.1 | 5/2, (7/2)+ |

^a This work.
^b Wagner *et al.*,¹⁴

1/2+, 5/2+, and 3/2+, respectively, constitute members of a common rotational band. The energies of levels in such a *K*=1/2 band are given by the formula³²

$$E_I = (\hbar^2/2\mathcal{I})[I(I+1) + a(-)^{I+1/2}(I+1/2)].$$

Solving for the two undetermined parameters, using the above energy levels, we obtain $\hbar^2/2\mathcal{I}=6.2$ keV and $a=+1.08$. The moment of inertia is about equal to that of the band based on the 60-keV state, where $\hbar^2/2\mathcal{I}=6.21$ keV.

Thus, the eight levels populated by the beta decay of U²³⁷ can be grouped into four bands: Band *A* based on the ground state has *K*=5/2+; Band *B* based on the 59.57-keV level has *K*=5/2-; Band *C* with one known level at 267.5 keV has *K*=3/2-; Band *D* based on the 332.3-keV level has *K*=1/2+.

On considering intensity relationships let us first look for transitions violating the *K*-selection rule ($\Delta K \leq L$, the multipolarity). The only ones are the *M*1 transitions of 335.3, 368.5, and 370.9 keV. The only important competing transitions should be the low-energy (38.6- and 36.2-keV) intra-band transitions to the 1/2+ state. A special search was made for lines corresponding to these possible transitions. Slight evidence was obtained for a 38.6-keV transition, namely an *L*_{II} line, but unfortunately the expected positions of *L*_{III}, *M*_I, and *M*_{II} are masked by lines of other transitions. The *L*_I line (not masked) is not seen. A 36.2-keV transition is not observed, no lines being found in *L*_{III} and *M*_{II} positions (not masked). The beta-continuum background on the plates is high in the region where the *L* and *M* lines of these possible transitions might be found, and the efficiency of the emulsion at these low energies is poor. Hence, it can only be said that any 36.2-keV transition must be less than about one-half percent of total beta decay, and the 38.6-keV transition may occur of the order of one percent of the beta decay if the observed *L*_{II} line does not arise from some other source. The very approximate nature of the above intensity information introduces a good deal of uncertainty into the question of the proportion of primary beta decay to the three levels of band *D*. The most we can say is that the total beta population to the band comprises about five percent. Within the uncertainty on possible gamma rays competing with the 335.3-, 368.5-, and 370.9-keV transitions a retardation associated with *K* forbiddenness is consistent.

³² A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd 27, No. 16 (1953).

³³ S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd 29, No. 16 (1955).

It is of interest to compare the observed gamma-ray branching ratios with the relative reduced transition probabilities calculated from the Bohr-Mottelson model; such comparisons are in effect tests of the purity of the K -quantum numbers of initial and final states. In the case of the two $E1$ transitions which de-excite the 59.6-keV state (26.4 and 59.6 keV) such a check has been made from Am^{241} alpha decay by HSR,¹ who found the theoretical and experimental ratios in disagreement by over a factor of two.

Consider now the branching of $E2$ radiation from the $3/2^-$ level (267.5 keV) to states of band B . As discussed earlier, there is very little $E2$ admixture in the 207.9-keV transition, the value based upon the L_{III} -conversion coefficient being $E2/M1=0.005\pm 0.005$. The intensity rules of Alaga *et al.*³¹ predict

$$[B_{208}(E2)/B_{165}(E2)]_{\text{theor}}=0.75,$$

but experimentally the ratio is between zero and 0.1. This extremely bad disagreement is surprising, and we cannot explain it in detail. As we shall show later, this $E2$ transition of the odd proton is hindered, probably because of a violation of a selection rule in the asymptotic quantum numbers. One might expect some contribution to the $E2$ radiation by virtue of Coriolis (RPC, see Kerman)¹⁰ admixing of the intrinsic proton wave

functions of bands B and C . Perhaps a destructive interference of these contributions has lowered the $E2$ component of the 207.9-keV gamma.

Consider also the $M2$ branching from the $3/2^-$ level to states of band A . The Clebsch-Gordan coefficients in the theoretical expressions are the same as for the $E2$ branching just considered. That is,

$$[B_{267}(M2)/B_{234}(M2)]_{\text{theor}}=0.75.$$

The experimental ratio of reduced transition probabilities appears to be

$$[B_{267}(M2)/B_{234}(M2)]_{\text{expt}}=3.3,$$

in bad disagreement. Possible explanations might be (1) an abnormally high K -conversion coefficient³⁴ for the $E1$ component of γ_{267} or (2) an unexpectedly large contribution from intrinsic states with $K\neq 5/2$ in the final state. The $E1$ K -conversion coefficient would need to be a factor of 12 larger than theoretical to bring the $M2$ branching ratio into agreement with theory.

Agreement with the relative intensity rules of Alaga *et al.*³¹ is not found in the three cases listed above, and in the Bohr-Mottelson formalism this means that in each case there are important contributions to the transition probability resulting from wave-function components with K values differing from that of the principal component.

Correlations Dependent on the Odd-Proton Wave Function

We shall now discuss the more special considerations involving the state of intrinsic motion of the odd proton in Np^{237} . Considerable success has been achieved in correlating ground-state spins and excited-nucleonic-state spins of deformed nuclei with the Ω values of nucleon eigenfunctions calculated by Nilsson³³ for the anisotropic harmonic-oscillator potential. We wish to associate the four bands of Np^{237} and the ground state of U^{237} with states from Nilsson's calculations. In testing possible assignments we shall examine both the selection rules in the asymptotic (large-deformation) quantum numbers N , n_z , and Λ and those in the spherical limit in j and l (and N). (See papers of Nilsson,³³ Alaga,³⁵ Hollander,³⁶ Perlman and Rasmussen.⁹) For convenient reference we show in Fig. 2 a diagram of the Nilsson eigenvalues of the heavy region for prolate deformation. Calculations from experimental quadrupole moments indicate a deformation of about ~ 0.25 . In the right-hand margin are the letters A , B , C , and D , adjacent to the states we associate with the four bands in Np^{237} . The reasons for these choices are the following:

³⁴ Anomalous large L -conversion coefficients have been observed experimentally from Am^{241} decay (reference 2) and Th^{231} decay (reference 30).

³⁵ G. Alaga, Phys. Rev. **100**, 432 (1955).

³⁶ J. M. Hollander, Phys. Rev. **105**, 1518 (1957).

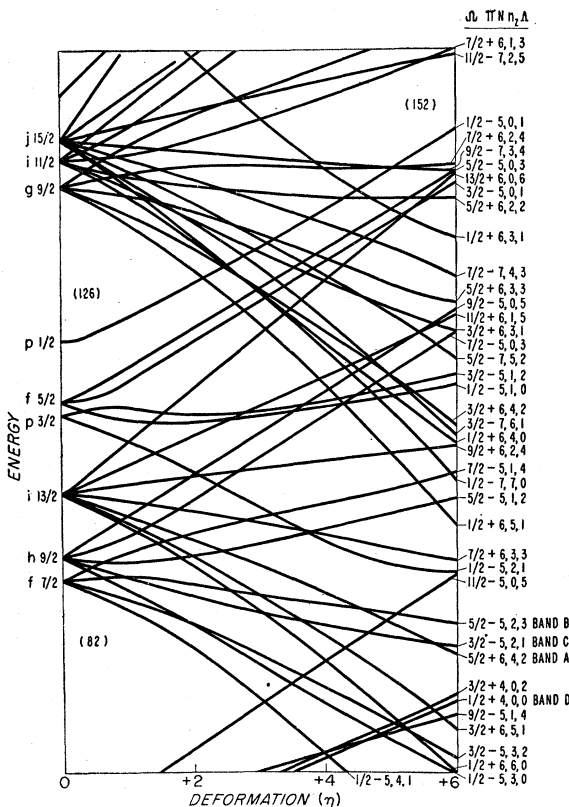


FIG. 2. Nilsson energy-level diagram for prolate deformation.

From the ground-state spins, rotational-band spacing, and the unhindered nature of one alpha-decay group in Am²⁴¹ it was suggested¹⁶ that the odd proton is in orbitals of $\Omega=5/2$ in the states both of band *A* and band *B*. These orbitals are necessarily of opposite parity because of their interconnection by electric dipole radiation. It was later suggested by HSR¹ that of the two available $\Omega=5/2$ orbitals in the proper region band *A* is the even-parity orbital, the chief evidence being the difference in effective moments of inertia of bands *A* and *B*. Magnetic-moment calculations of Strominger³⁷ also support the above assignments.

From the nature of the four transitions depopulating the 267.5-keV level only the assignment $3/2^-$ seems reasonable, and the level labelled "C" in Fig. 2 is the only $3/2^-$ state in this region.

The $1/2^+$ band *D* would not be expected to lie this low in energy if one were to take strictly the eigenvalues of Nilsson, but taking Nilsson's diagram as simply a good guide to the position of states we shall examine in more detail the properties of the two nearest $1/2^+$

TABLE X. Partial half-lives for photon emission from the 267.5-keV state.

| Energy | Multi-polarity treated | Experimental half-life (sec) | Moszkowski single-particle estimate (sec) | Retardation factor F |
|--------|------------------------|------------------------------|---|------------------------|
| 267.5 | E1 ^a | 7.7×10^{-7} | 3.5×10^{-15} | 4.5×10^{-9} |
| 234.2 | M2 | 6.0×10^{-6} | 8.3×10^{-8} | 1.4×10^{-2} |
| 207.9 | M1 ^a | 2.0×10^{-8} | 1.5×10^{-12} | 7.5×10^{-5} |
| 164.6 | E2 | 1.6×10^{-7} | 9.5×10^{-9} | 5.9×10^{-2} |

^a These transitions have some quadrupole admixture, but the experimental partial lifetime applies to the dipole component alone.

orbitals on the diagram (the notation is $\Omega\pi\{N, n_z, \Lambda\}$). These are state $1/2^+ \{4, 0, 0\}$ and state $1/2^+ \{6, 6, 0\}$. Using Nilsson's wave functions for the former, we calculate at $\delta = +0.2$ a value $a = +1.29$ and at $\delta = +0.3$ a value $a = -0.67$. For the latter, we calculate $a_{+0.2} = +6.6$ and $a_{+0.3} = 6.2$. Since experimental quadrupole moments indicate $\delta \approx +0.25$ in the region of Np²³⁷ we feel that the agreement between the experimental $a = +1.08$ and the theoretical decoupling parameters for the $1/2^+ \{4, 0, 0\}$ orbital is satisfactory and that the $1/2^+ \{6, 6, 0\}$ orbital is conclusively ruled out.

A curious case is provided by the four gamma transitions depopulating the 267.5-keV level in that *E1*, *M1*, *E2*, and *M2* transitions are all observed. The lifetime of the parent state has been measured by Bunker *et al.*¹⁸ as $(5.4 \pm 0.5) \times 10^{-9}$ sec. In Table X we have made use of this lifetime measurement and our relative intensity data to calculate experimental partial half-lives for emission of the various photons. For a theoretical comparison we have made calculations also with use of the single-proton transition formulas of

³⁷ D. Strominger, University of California Radiation Laboratory Report, UCRL-3374, June 1956 (unpublished).

TABLE XI. Asymptotic selection rules for gamma transitions.

| ΔN | Δn_z | $\Delta \Lambda$ | $\Delta \Sigma$ | ΔN | Δn_z | $\Delta \Lambda$ | $\Delta \Sigma$ |
|------------|--------------|------------------|-----------------|----------------|--------------|------------------|-----------------|
| <i>E1</i> | | | | <i>M1</i> | | | |
| +1 | +1 | 0 | 0 | 0 | 0 | 0 | ± 1 |
| -1 | -1 | 0 | 0 | 0, ± 2 | ± 1 | ± 1 | 0 |
| ± 1 | 0 | ± 1 | 0 | 0, ± 2 | 0 | 0 | 0 |
| <i>E2</i> | | | | <i>M2</i> | | | |
| +2 | +2 | 0 | 0 | $\pm 1, \pm 3$ | ± 1 | 0, ± 2 | 0 |
| -2 | -2 | 0 | 0 | $\pm 1, \pm 3$ | 0 | ± 1 | 0 |
| 0, ± 2 | 0 | 0 | 0 | $\pm 1, \pm 3$ | ± 2 | ± 1 | 0 |
| 0, ± 2 | ± 1 | ± 1 | 0 | ± 1 | 0 | ± 1 | ± 1 |
| ± 2 | 0 | ± 2 | 0 | ± 1 | ± 1 | 0 | ± 1 |

Moszkowski.³⁸ None of these transitions is *K*-forbidden. The retardation of the *E1* is not too surprising, since the low-energy *E1* transitions quite generally show large retardation. The general question of retardation of *E1* transitions in deformed nuclei has been discussed by Strominger and Rasmussen.³⁹

The large retardation of the 208-keV *M1* transition, allowed by *K*-selection rules, needs explanation. A qualitative explanation is readily found with the earlier orbital assignments by examining selection rules in the asymptotic quantum numbers. Selection rules in the quantum numbers *N* (total-oscillator quantum number), *n_z* (*z*-axis oscillator quantum number), Λ (orbital angular-momentum component along symmetry axis), and Σ (projection of intrinsic spin along symmetry axis)⁴⁰ have been applied to cases of beta decay by Alaga³⁵ and to cases of *E1* gamma transitions by Strominger and Rasmussen.³⁹ Chasman and Rasmussen⁴¹ have derived selection rules for *E1*, *E2*, *M1*, and *M2* transitions. These selection rules are given in Table XI.

From the asymptotic quantum numbers appropriate to orbitals *C* and *B* in Fig. 2 and from inspection of Table XI it is seen that the *M1* transition violates these rules, since $\Delta \Lambda = 2$.

Table X shows the 164.6-keV *E2* as retarded, and the *E2* component of the 207.9-keV transition must be even more retarded. It is to be noted that an *E2* single-proton transition between the *B* and *C* orbitals violates the selection rule $\Delta \Sigma = 0$. The speed of the *E2* transitions between these bands is a more difficult matter to consider theoretically since the Coriolis interaction (Kerman's¹⁰ RPC interaction) will mix these proton states and lead to collective contributions to the *E2* transition probabilities. The single-particle and collec-

³⁸ S. A. Moszkowski, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955), Chap. XIII.

³⁹ D. Strominger and J. O. Rasmussen, Nuclear Phys. **3**, 197 (1957).

⁴⁰ Σ may take only the values $\pm 1/2$. This is not explicitly given in Fig. 2 but is easily determined since $\Omega = \Lambda + \Sigma$.

⁴¹ R. Chasman and J. O. Rasmussen, University of California Radiation Laboratory Chemistry Division Quarterly Report, January, 1957 (unpublished).

tive contributions could interfere constructively or destructively; the latter possibility is attractive for explaining the retarded $E2$ component of the 207.9-keV gamma.

We see from Table X that the $M2$ radiation is also retarded from the single-particle estimate, but it is significantly less retarded than the $M1$. $M2$ radiation between bands C and A is allowed by the asymptotic quantum-number selection rules of Table XI.

Finally, let us consider the beta decay $\log ft$ values (Table IX) and attempt to assign quantum numbers to the ground state of U^{237} . To account for the non-observance of beta decay to the $5/2+$ and $5/2-$ bands a spin assignment of $1/2$ seems most attractive. The available $\Omega=1/2$ neutron states in the proper region on Nilsson's diagram are $1/2+ \{6,3,1\}$ and $1/2- \{5,0,1\}$. For the even-parity assignment the principal beta group to the $3/2-$ state would be classified first forbidden ($\Delta I=1$, yes), unhindered. The experimental $\log ft$ value of 6.2 seems quite consistent with this from comparison with Alaga's study.³⁵ For the odd-parity assignment the main beta group would be allowed ($\Delta I=1$, no), hindered, also consistent with experiment.

Although there is much uncertainty regarding the

relative beta intensities to the uppermost band, a self-consistent picture explaining beta decay to this upper band is more easily obtained with the assignment of $1/2-$ to U^{237} . The beta decay to the $1/2+$ and $3/2+$ states would then be classified first forbidden, unhindered, and beta decay to the $5/2+$ state would be first forbidden ($\Delta I=2$, yes), unhindered. The $1/2+$ assignment to U^{237} would not allow sufficient direct (second forbidden) beta population of the $5/2+$ state at 368.5 keV, and it would be necessary to postulate a higher level decaying to it by an unobserved gamma transition.

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