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# THE DECAY OF NEPTUNIUM-238

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ABSTRACT

A study has been made of the energy levels of $Pu^{238}$, which are populated by $Np^{238}$ beta decay, by an examination of the $Np^{238}$ conversion electron spectrum in high-resolution beta spectrographs. Agreement is found with the general features of the decay scheme as previously given, but three new transitions are observed and placed in the scheme. These have energies of 885-, 943-, and 988-kev. The formerly unresolved transitions of ~ 1030-kev have been resolved into two components, 1027-kev and 1030-kev. Comparisons are made of the experimental relative transition intensities with those predicted from the rules of Alaga et al., for transitions depopulating the beta and gamma vibrational bands, and satisfactory agreement is obtained providing the postulate is made that the 940 and 943 kev transitions are of the electric monopole type.

The beta decay log ft values are consistent with spin and parity values of 3, even or odd, or 2, odd for ground state of $Np^{238}$.

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## Introduction

The energy levels of $Pu^{238}$ have been studied extensively from the decay[1-3] of $Cm^{242}$ and the decay[4-8] of $Np^{238}$. The alpha decay of $Cm^{242}$ leads to a well-defined ground state rotational band, a state of 605 kev which has been assigned[2] as (0,1-),[9] and two states of 937 kev and 1030 kev. The high energy states of $Pu^{238}$ are populated by only a very small fraction of the total alpha decay and (selection rules permitting) are studied more easily through the beta decay of $Np^{238}$. The spectroscopic studies of this nuclide by Rasmussen et al.[4,5] and by Baranov and Shlyagin[8] have defined levels at about 988-, 1030-, and 1076-kev; these levels are of theoretical interest because of their tentative interpretation[1,5] as vibrational states.

Since in the earlier experimental studies the two prominent transitions of about 1030 kev energy had never been completely resolved, it had not been possible to correlate fully the experimental transition intensities with those predicted from considerations of the Bohr-Mottelson model.[10,11] For this reason we have re-examined the conversion electron spectrum of $Np^{238}$ on the high-resolution permanent-magnet electron spectrographs of this laboratory. It was hoped also that the transition between the (2,2+) and the (0,4+) states might be found, so that its intensity could be compared with the theoretically expected value.

Sample Preparation

The $Np^{238}$ sample was prepared by a 5-hour neutron irradiation of approximately 200 micrograms of $Np^{237}$ in the Materials Testing Reactor at Arco, Idaho.

The sample was dissolved in concentrated HCl, and the neptunium coprecipitated with $Zr_3(PO_4)_4$ after reduction to the +4 state by $Fe^{++}$. Further purification was effected by co-precipitations with $LaF_3$ and $La(OH)_3$. To separate the neptunium from lanthanum and other rare earths, the solution was passed through a Dowex A-1 anion column at high chloride concentration and the neptunium subsequently eluted with 1 M HCl. This sample was electroplated onto a 10 mil platinum wire which served as the source for the spectrographs. Used in the experiment were three photographic recording permanent magnet spectrographs with field strengths of 50, 100, and 340 gauss and resolving powers ($\Delta\rho/\rho$) of approximately 0.1%.[3]

Experimental Results

The experimental results are summarized in Table I. The reported transition energies ($E_\gamma$) are weighted averages of the results of several exposures. Since no internal standard was used, absolute errors are not given; however, except for very weak lines the relative precision of the energies should be ~ 0.1%. The value 44.11 kev listed in parentheses has been used in the decay scheme instead of the value 44.00 kev obtained in this experiment, because the former value was obtained with use of several internal standards,[3] and is considered the more accurate.

We have attempted to summarize in Fig. 1 the presently known information about the $Pu^{238}$ level scheme, including the $Np^{238}$ beta decay and $Cm^{242}$ alpha decay data. This scheme is similar to those given by Rasmussen et al.[5] and by Baranov and Shlyagin,[8] with small differences in the energies of some of the states. Included are three new transitions seen in the present work (885-, 943-, and 988-kev) and also the levels at 514-, 605-, and 935-kev which have not been observed from $Np^{238}$ beta decay but were found by Asaro et al.[2] from $Cm^{242}$ alpha decay. The transition intensities shown in the scheme are those deduced from a consideration of our electron intensities together with previous information on the beta decay branchings; these data will be discussed below.

Experimental relative intensities of the conversion lines were difficult to obtain by densitometry due to the beta background on the photographic plates. The relative intensities listed in Table I are the results of six visual estimates that were made by comparing the conversion lines with intensity standards. Corrections were made for instrument geometry[3,12] and film efficiency. For purposes of comparison the intensities are normalized to a value of 0.20 for the K line of the 985.7 kev transition. They are probably accurate to about 25% for the stronger lines and to a factor of 2 for the weaker lines. Intensities reported by the other investigators are also listed for comparison. Note that our intensity values for the components of the ~ 1030 kev doublet, which are completely resolved, disagree with the results of Baranov and Shlyagin;[8] we observe that the higher energy (1030 kev) component is more intense than the lower energy component (1027 kev) whereas Baranov and Shlyagin report the opposite result. The other investigators reported a composite intensity without attempting to divide it between the two transitions.

Branching-Ratio Comparisons

It is of interest to compare the experimental transition intensities with theoretical values calculated from the rules of Alaga, Alder, Bohr, and Mottelson[11] which state that the reduced transition probabilities of photons of the same multipole order from a given initial state to different final states within a rotational band are proportional to the squares of the vector addition coefficients connecting these states. These rules follow from the general features of the Bohr-Mottelson model, and their validity depends upon the purity of the K quantum number. The vector addition coefficient for a transition of multipole order L from an initial state i to a final state f is given in the representation $\langle I_i \; L \; K_i \; (K_f - K_i) \mid I_i \; L \; I_f \; K_f \rangle$. Numerical values of these coefficients have been tabulated by Simon.[13]

From the experimental K-conversion coefficients measured by Rasmussen et al.[4,5] and by Baranov and Shlyagin[8] it is fairly certain that the 1030- and 986-kev transitions are predominantly electric quadrupole. It has also been inferred that the 940 and 925 kev transitions are E2.

We have calculated the theoretical relative K-electron intensities from the theoretical relative reduced E2 photon transition probabilities, given by the vector addition coefficients, by correcting for the 5th power energy dependence and multiplying by the theoretical K-conversion coefficients.[14] The experimental and theoretical electron intensities are compared in Table II. Because of the large uncertainties in the experimental electron intensity figures, quantitative comparison of experimental and theoretical intensities is not justified; however, it appears that for the three transitions which depopulate the 1030 (2,2+) level there is good agreement between theory and experiment. This agreement tends to confirm the K = 2 assignment[5] of this band and to indicate a fairly high degree of K-purity. On the other hand, the ratio of the experimental transition intensities from the 1071 (2,3+) level differs from the theoretical value by almost a factor of three. Because it is so difficult to estimate error limits on these intensities, we are not certain that this disagreement is real.

## Discussion of Other Transitions

In addition to the transitions considered in the preceding section, four others were observed in this work; these have energies 940-, 943-, 988-, and 871-kev. Only the 940-kev has been reported previously. The 871-kev transition may not be real because its "K-line" was extremely weak and uncertain; we shall not discuss this transition further.

There is insufficient information to fit the remaining three transitions uniquely into the level scheme. We shall here examine two alternative possibilities which can be postulated. One of these two seems more plausible, but final choice must await further experiment.

Previous investigations[2,5,8] have defined levels at approximately 937 and 988 kev in addition to those already discussed and it has been suggested[1] that these are the first two members of a beta vibrational band, with K = 0. Such vibrational levels are predicted by the unified model of Bohr and Mottelson[10] and are expected to occur at an excitation energy of about 1 Mev in the heaviest elements. Consider the observed transitions of 940-, 943-, and 988-kev in relation to these (0,0+) and (0,2+) levels.

The two alternative possibilities to be discussed are shown in Fig. 2. In Fig. 2a the 940-kev transition is assumed to be the same as the transition of 937-kev observed from $Cm^{242}$ alpha decay (the energy difference is within experimental error). The 0,0+ state is thus defined at ~ 940 kev. The 943 and 988 kev transitions, whose energy difference approximates the energy of the first excited state, are placed so as to de-excite a 0,2+ level at 988 kev. But in spite of this energy sum, there are intensity considerations which make assignment 2a unlikely. These concern the relative strengths of electric quadrupole and electric monopole transitions expected from the 0,2+ level. Perlman and Asaro[16] have observed, from $Cm^{242}$ alpha decay, that the E0 transition from the 0,0+ level to ground (~ 937 kev) is about one-half as strong as the E2 transition (~ 890 kev) from this level to the 0,2+ state at 44 kev. Because of the great difference in conversion coefficients ($e_K/\gamma$ (E2) ~ 1%, $e_K/\gamma$ (E0) = $\infty$) the K-line of the E0 transition is 50 times stronger than that of the E2 transition. Such an observation is consistent with the theoretical considerations of Church and Weneser[15] concerning E0 transition probabilities.

If it is reasonable to assume that such competition between E0 and E2 transitions would be similar in the case of de-excitation of the 988 kev (0,2+) level, then one would expect the K-line of the 943-kev transition to be about a hundred-fold stronger than that of the 988-kev transition. Instead, we find them to be about equal in intensity. We conclude that situation 2a is probably not correct, and that the energy sum 44 + 943 $\approx$ 988 is fortuitous.

In the second alternative, Fig. 2b, the moderately strong 940-kev transition is assigned as the electric monopole transition which proceeds between the two 0,2+ levels. This assignment leads to a log ft value for beta decay to the 985-kev (0,2+) level similar to that of the observed beta group which populates the 44-kev (0,2+) level (see beta decay section). Assignment of the 943 and 988 kev transitions can now be made only by postulating new levels. We shall do this, within the framework of the K = 0 and K = 2 character of the high-lying states as discussed above.

From the energy spacings between the ground and first excited states of each of these bands one obtains the value of the rotational constant $\hbar^2/2\mathfrak{J}$ from the formula[17]

$$E_{rot} = \hbar^2/2\mathfrak{J} \; [\, I\,(I + 1) - I_o\,(I_o + 1)\,].$$

These values are found to be: 7.35 kev for the ground state band, 8.0 kev for the beta (K = 0) vibrational band, and 6.83 kev for the gamma (K = 2) vibrational band. From these rotational constants one calculates the energies of the next higher levels of the beta (0,4+) and gamma (2,4+) vibrational bands to be 1097- and 1126-kev, respectively. We shall postulate the following assignments: the 943 kev transition takes place between the upper and lower (0,4+) levels, and the 988 kev transition takes place between the (2,4+) and lower (0,4+) levels. Thus, we define experimental energies of the (0,4+) and (2,4+) states as 1089- and 1134-kev, respectively. The energy difference between these two experimental values is greater than that between the calculated energies, but such a deviation could be caused by configuration interaction between the two 4+ levels.

The 943-kev transition depopulating the proposed (0,4+) level can, from the previous arguments, be expected to have a major E0 component. This is consistent with the fact that it is the only transition seen from this level in the conversion-electron spectrum.

By analogy with the other levels of the K = 2 band, one expects the (2,4+) state to de-excite by E2 transitions. (Since in this band K = 2, electric monopole transitions to states of the K = 0 band are forbidden by the K-selection rule.) The theoretical E2 branching ratios, as given in Table II, predict that the (2,4+) ⟶ (0,4+) transition should be twice as strong (in the electron spectrum) as the stronger of the other two possible transitions. Experimentally it is the only one seen, and since it was just barely detectable any weaker transitions would have been missed. It would be desirable, with more intense sources of $Np^{238}$, to attempt to find and determine the intensities of the (2,4+) ⟶ (0,2+) and (2,4+) ⟶ (0,6+) transitions, which should appear at 1090- and 830-kev respectively.

## $Np^{238}$ Log ft Values and Spin

We shall consider finally the beta decay branchings and spin of $Np^{238}$. Information regarding the total intensity of the soft-beta component has been obtained by Freedman et al.,[6] Rasmussen et al.,[4] and by Baranov and Shlyagin;[8] their respective values, in fairly good agreement, are 53%, 55%, and 59%. In order to obtain intensities of the individual transitions, we convert our measured (relative) electron intensities to transition intensities by use of Sliv's theoretical E2 conversion coefficients[14] and then place these on an absolute scale by requiring that the total equal 56% (average of above determinations). The values so obtained are compared in Table III with those obtained directly from the absolute electron intensity measurements of Slätis et al.[7] and of Rasmussen et al.[4] The agreement is good.

From the above information, log ft values of beta decay to the various levels were calculated, and are given in Table IV. It is to be emphasized that these calculations are not independent of the present assignment of the multipole orders of the various transitions; however, the only ft values which are affected strongly are those of the beta decay to the 985- and 1089-kev levels. If the transitions which de-excite these states should prove to be Ml's instead of E0's, then the log ft values will decrease by one unit.

On the basis of the present data in appears that the spin of $Np^{238}$ is 3, with even or odd parity, or possibly 2 with odd parity. The spin 3 assignment explains the failure to observe beta decay to the two (0,0+) levels and to the (0,1-) level of $Pu^{238}$, and it is also consistent with the log ft values 6.2, 6.5, and 6.9 of beta decays to the (2,2+), (2,3+), and (2,4+) levels, respectively. Because these log ft values seem a little low for transitions of the $\Delta$ I = 1, yes type, we favor the assignment of even parity to $Np^{238}$. With this choice, the beta decays to the 2+ and 4+ levels of the two K = 0 bands are of the "allowed" type by ordinary $\Delta$ I selection rules. That they are all highly retarded (log ft's 8.5 - 9.4) is interpreted as arising from the serious violation of the K-selection rule which occurs, since in these transitions $\Delta$ K exceeds $\Delta$ I by two units. Possible evidence against the spin 3 assignment comes from the uneven branching of the beta decays to the 2+ and 4+ states of the ground band. It might be expected that such transitions would have similar log ft values; instead the 2+ final state is heavily favored over the 4+ final state. With the alternative choice of spin and parity 2, odd, it is not easy to compare the ft values of these two beta transitions because one of them would be K-forbidden and the other would not. On the other hand, if our postulated (2,4+) level is correct, a spin value of 2 is contraindicated since the observed log ft value for decay to that level (6.9) is much too small for a $\Delta$ I = 2 yes transition.

The assignment of 2, even, to $Np^{238}$ is ruled out by the observation by Baranov and Shlyagin[8] of beta population to the 146 kev (0,4+) level and also by the present tentative observation of decay to the 1134 kev (2,4+) level.

A direct measurement of the spin of $Np^{238}$ would be most desirable.

Our thanks are due Professor J. O. Rasmussen for suggesting the experiment and for many comments. We appreciate also the comments of Professor I. Perlman, Dr. F. Asaro, and Dr. F. S. Stephens. This work was performed under the auspices of the U. S. Atomic Energy Commission.

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Table I

$Np^{238}$ Conversion Electron Data

| Electron energy (kev) | Conversion shell | Transition energy (kev) (a) | Selected transition energy (kev) (b) | Abundances per 100 beta disintegrations (c) Freedman et al. | (d) Slätis et al. | (e) Rasmussen et al. | Relative (g) intensities (f) Baranov and Shylagin | Present work |
|---|---|---|---|---|---|---|---|---|
| 21.76 | $L_{II}$ | 44.01 | | 38 | 28.7 | -- | 23.1 | -- |
| 25.96 | $L_{III}$ | 44.02 | | 20 | 21.0 | -- | 21.1 | -- |
| 38.44 | $M_{II}$ | 44.00 | | 14 | | -- | 11.6 | -- |
| 39.40 | $M_{III}$ | 43.96 | | | | -- | 5.72 | -- |
| 42.58 | $N_{II}$ | 43.96 | | | 15.2 | -- | 3.71 | -- |
| 42.84 | $N_{III}$ | 43.97 | | 3.2 | | -- | | -- |
| 43.71 | O | ~44.0 | | | | -- | -- | -- |
| | | | (44.11)[(h)] | | | | | |
| 79.52 | $L_{II}$ | 101.8 | | 1.9 | 1.4 | -- | 1.21 | -- |
| 83.74 | $L_{III}$ | 101.8 | | | 0.9 | -- | 0.64 | -- |
| 96.20 | $M_{II}$ | 101.8 | | | | -- | 0.40 | -- |
| 97.17 | $M_{III}$ | 101.7 | | 1.2 | 0.7 | -- | 0.36 | -- |
| 100.3 | $N_{II}$ | 101.7 | | -- | -- | -- | -- | -- |
| 101.4 | O | 101.7 | | -- | -- | -- | -- | -- |
| | | | 101.7 | | | | | |
| 748.8 | K(?) | 870.6 | 870.6(?) | -- | -- | -- | -- | extremely weak |
| 762.8 | K | 884.6 | 884.6 | -- | -- | -- | -- | 0.016 |
| 803.6 | K | 925.4 | 925.4 | -- | 0.05 | 0.04 | -- | 0.036 |
| 818.8 | K | 940.6 | | -- | 0.10 | 0.07 | 0.09 | 0.076 |
| 917.9 | $L_I, L_{II}$ | 940.2 | | -- | 0.06 | -- | -- | 0.022 |
| 934.4 | $M_I, M_{II}$ | 940.0 | | -- | -- | -- | -- | 0.0072 |
| | | | 940.4 | | | | | |

(continued)

Table I (continued)

| Electron energy (kev) | Conversion shell | Transition energy (kev) (a) | Selected transition energy (kev) (b) | Abundances per 100 beta disintegrations (c) Freedman et al. | (d) Slätis et al. | (e) Rasmussen et al. | Relative (g) intensities (f) Baranov and Shylagin | Present work |
|---|---|---|---|---|---|---|---|---|
| 821.5 | K | 943.3 | 943.3 | -- | -- | -- | -- | 0.0098 |
| 864.0 | K | 985.8 | | 0.3 | 0.26 | 0.20 | 0.20 | 0.20 |
| 963.1 | $L_I,L_{II}$ | 985.4 | | } -- | } 0.13 | } 0.06 | 0.06 | 0.054 |
| 967.8 | $L_{III}$ | 985.9 | | | | | -- | 0.0092 |
| 980.2 | $M_I,M_{II}$ | 985.8 | | -- | -- | -- | -- | 0.018 |
| 985.3 | N | ~986 | | -- | -- | -- | -- | 0.0082 |
| | | | 985.7 | | | | | |
| 866.5 | K | 988.3 | 988.3 | -- | -- | -- | -- | 0.012 |
| 905.6 | K | 1027.4 | | 0.3 | 0.22 | 0.20 | 0.16(i) | 0.046 |
| 1005.1 | $L_I,L_{II}$ | 1027.4 | | -- | 0.08 | 0.04 | 0.03 | 0.016 |
| 1021.7 | M | ~1027 | | | 0.06 | 0.02 | -- | extremely weak |
| | | | 1027.2 | | | | | |
| 908.1 | K | 1029.9 | | -- | -- | -- | 0.08 | 0.14 |
| 1007.3 | $L_I,L_{II}$ | 1029.6 | | -- | -- | -- | 0.03 | 0.036 |
| 1024.8 | M | ~1030 | | -- | -- | -- | -- | 0.010 |
| | | | 1029.9 | | | | | |

(a) Electron binding energies were taken from Hill, Church, and Mihelich, Revs. Sci. Inst. 23, 523 (1952).

(b) The selected values are weighted averages of the experimental values.

(c) Reference 6 (d) Reference 7 (e) Reference 4 (f) Reference 8

(g) For purposes of comparison, the relative intensities of Baranov and Shylagin and of the present work listed here were normalized to Rasmussen's value of 0.20 for the K line of the 985 kev transition.

(h) The weighted average is 44.00. The value 44.11 had been reported previously by Smith and Hollander (Reference 3) and is considered more accurate.

(i) The intensities listed for the 1027 kev transition by Freedman, Slätis, and Rasmussen include those of the unresolved 1030 kev transition. The division of intensities by Baranov and Shlyagin between 1027 and 1030 is given as only approximate.

Table II

Theoretical and Experimental E2 K-Electron Intensity Ratios

| Transition ratio | Energy ratio | Ratio of[a] theoretical E2 reduced photon transition probabilities | Ratio of[b] theoretical E2 K-electron intensities | Ratio of experimental K-electron intensities |
|---|---|---|---|---|
| $\frac{(2,3+)\rightarrow(0,4+)}{(2,3+)\rightarrow(0,2+)}$ | $\frac{925}{1027}$ | 0.40 | 0.28 | 0.78 |
| $\frac{(2,2+)\rightarrow(0,0+)}{(2,2+)\rightarrow(0,2+)}$ | $\frac{1030}{986}$ | 0.70 | 0.74 | 0.71 |
| $\frac{(2,2+)\rightarrow(0,4+)}{(2,2+)\rightarrow(0,2+)}$ | $\frac{885}{986}$ | 0.050 | 0.069 | 0.079 |
| $\frac{(2,4+)\rightarrow(0,2+)}{(2,4+)\rightarrow(0,4+)}$ | $\frac{1090}{988}$ | 0.34 | 0.47 | --- |
| $\frac{(2,4+)\rightarrow(0,6+)}{(2,4+)\rightarrow(0,4+)}$ | $\frac{830}{988}$ | 0.086 | 0.050 | --- |

a. i.e. ratio of squares of vector addition coefficients.

b. Theoretical K-electron intensities were obtained by correcting the theoretical reduced photon transition probabilities for energy dependence and conversion coefficients.

Table III. Intensities of High-Energy Transitions in $Np^{238}$ Decay

| $E_\gamma$ (kev) | K-electron intensities (percent of disintegrations) Slätis[b] | Rasmussen[c] | Present work | $\alpha_K$[d] (Theoretical) | Transition intensity (percent of disintegrations) Slätis[b] | Rasmussen[c] | Present work |
|---|---|---|---|---|---|---|---|
| 871 | -- | -- | --[a] | | -- | -- | --[a] |
| 885 | -- | -- | 0.019 | $1.5 \times 10^{-2}$ | -- | -- | 1.6 |
| 925 | 0.05 | 0.04 | 0.042 | $1.05 \times 10^{-2}$ | 5 | 4 | 4.1 |
| 940 | 0.10 | 0.07 | 0.09 | $\infty$ | 0.10 | 0.07 | 0.09 |
| 943 | -- | -- | 0.01 | $\infty$ | -- | -- | 0.01 |
| 986 | 0.26 | 0.20 | 0.24 | $1.03 \times 10^{-2}$ | 26 | 20 | 23 |
| 988 | -- | -- | 0.014 | $0.99 \times 10^{-2}$ | -- | -- | 1.4 |
| 1027 | {0.22} (1027 and 1030) | {0.20} (1027 and 1030) | 0.054 | $0.88 \times 10^{-2}$ | {22} (1027 and 1030) | {20} (1027 and 1030) | 6.2 |
| 1030 | | | 0.17 | $0.88 \times 10^{-2}$ | | | 19 |

a. K-line seen, but intensity was too low to measure.

b. Reference 7

c. Reference 4

d. Reference 14

Table IV. $Np^{238}$ log ft Values

| Final state (K, I, π) | Final state (kev) | $E_{\beta^-}$ [a] | Percent of beta decay | log ft |
|---|---|---|---|---|
| 0, 0+ | 0 | (1290) | -- | -- |
| 0, 2+ | 44.11 | 1246 | 42 | 8.5 |
| 0, 4+ | 145.8 | 1144 | ~3 | 9.4 |
| 0, 6+ | 304 | (986) | -- | -- |
| 0, 8+ | 514 | (776) | -- | -- |
| 0, 1- | 605 | (685) | -- | -- |
| 0, 0+ | 937 | (353) | -- | -- |
| 0, 2+ | 985 | 305 | 0.09 | 9.1 |
| 2, 2+ | 1030 | 260 | 44 | 6.2 |
| 2, 3+ | 1071 | 219 | 10 | 6.5 |
| 0, 4+ | 1089 | 201 | 0.01 | 9.4 |
| 2, 4+ | 1132 | 158 | 1.4 | 6.9 |

a. Energies of beta transitions are obtained by subtraction of the energy of the particular level from the total decay energy, 1290 kev. Those transitions in parenthesis are not observed from $Np^{238}$ decay.

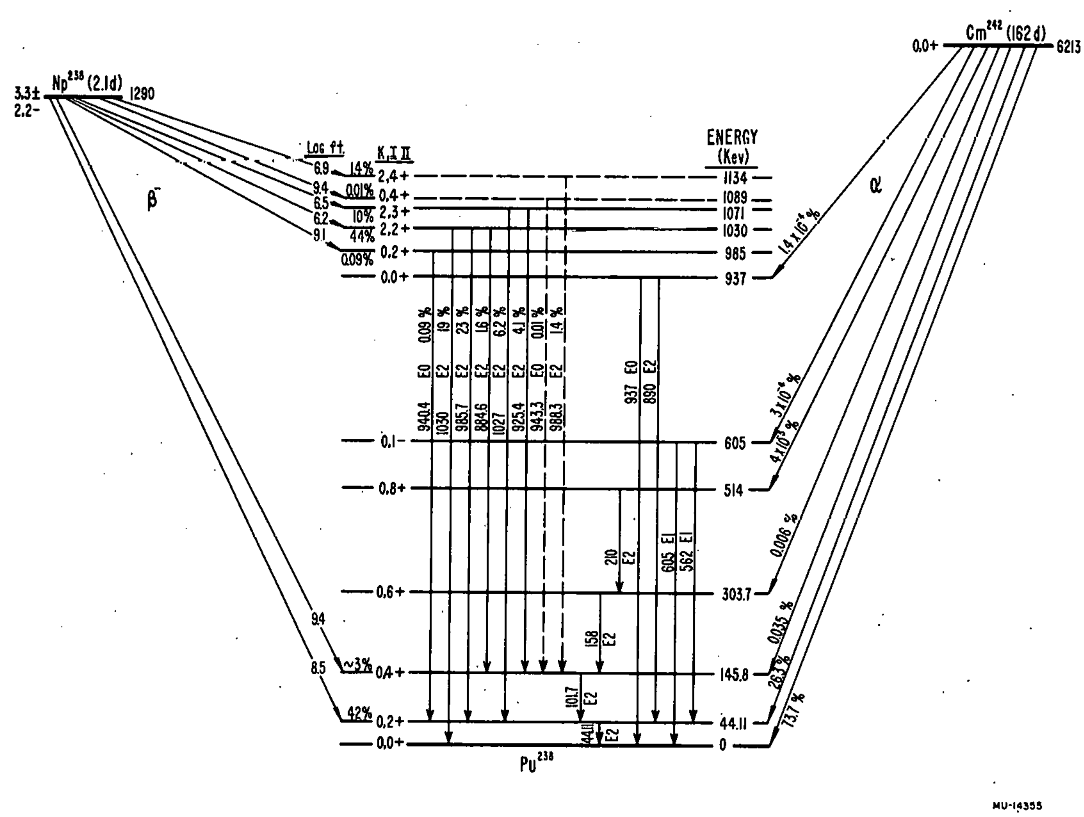


Fig. 1 Energy levels of $Pu^{238}$

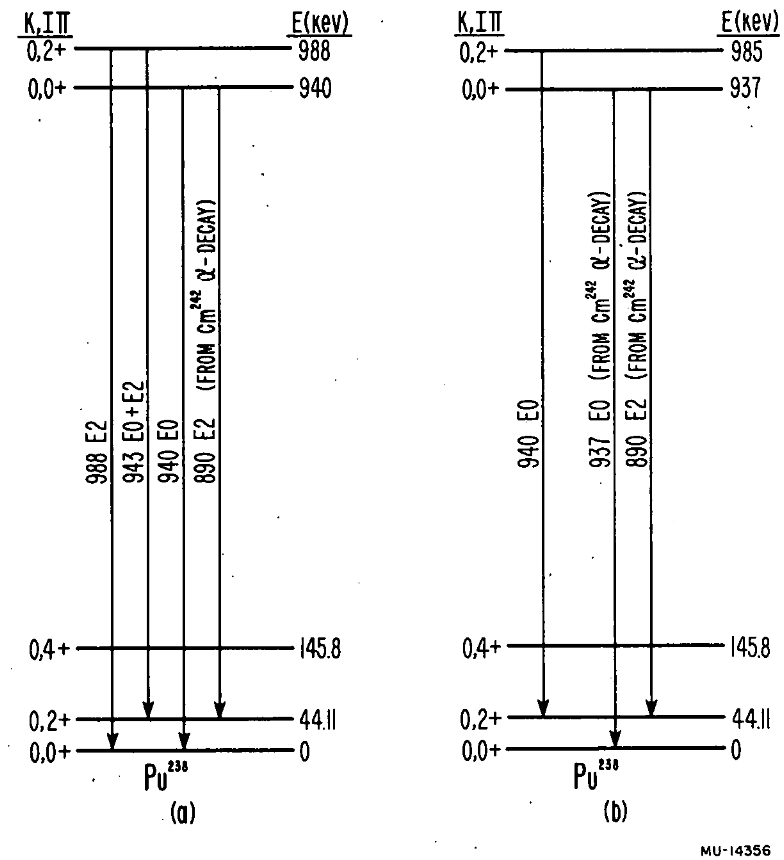


Fig. 2 Possible assignments of the 940-, 943-, and 988-kev transitions.