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February 1969

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A LOW-LYING 2- STATE IN  $^{236}\text{U}^\dagger$ 

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## Abstract

A low-lying  $KI\pi = 22-$  state of  $^{236}\text{U}$ , first characterized in measurements of  $^{240}\text{Pu}$  alpha decay, has been more thoroughly studied in the electron-capture decay of  $^{236}\text{Np}$ . Its energy is  $687.70 \pm 0.10$  keV, its half-life is  $(4.4 \pm 0.6) \times 10^{-9}$  seconds, and it decays by M2 and E1 transitions to members of the ground-state band. It is populated with an intensity of  $1.5 \pm 0.3\%$  in the decay of  $^{236}\text{Np}$ , and  $(2.1 \pm 0.4) \times 10^{-5}\%$  in the decay of  $^{240}\text{Pu}$ , in the latter case presumably via an alpha transition to the unobserved spin-3 member of the band. Besides having the lowest energy of any known 2- state in an even nucleus, the state is characterized by fast M2 transitions and highly hindered E1 transitions.

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## KEYWORDS

E RADIOACTIVITY  $^{236}\text{Np}$ ,  $^{240}\text{Pu}$ ,  $^{97}\text{Nb}$ ; measured  $E_{\gamma}$ ,  $I_{\gamma}$ ,  $E_{ce}$ ,  $cc$ ,  
 $E_{\beta^-}$ ,  $I_{\beta^-}$ , KX  $\gamma$ -delay,  $T_{1/2}$ ,  $\alpha$   $\gamma$ -,  $\alpha$   $ce$  coin;  $^{236}\text{U}$ ,  $^{236}\text{Pu}$ ,  
deduced levels,  $I\pi$ , log ft.

## 1. Introduction

The present investigation arose from apparent inconsistencies observed in  $\alpha$ - $\gamma$  and  $\alpha$ - $e^-$  coincidence studies of  $^{240}\text{Pu}$  decay<sup>1</sup>). Those measurements, made in 1962-1963 showed that  $\alpha$  decay of  $^{240}\text{Pu}$  populates a state around 693( $\pm$ 20) keV excitation in  $^{236}\text{U}$ . Gallagher and Thomas had previously observed transitions of 643 and 688 keV in the decay of  $^{236}\text{Np}$ , which they assigned as transitions from a 688 keV state to the ground and first-excited states of  $^{236}\text{U}$  (ref.<sup>2</sup>); it seemed likely that this was indeed the same state.

Systematics of level energies and  $\alpha$ -hindrance factors<sup>1</sup>) would suggest that this is the lowest member of the  $K = 0$  octupole-vibrational band. However, the conversion-electron data obtained from the coincidence measurements, described in sec. 2, seemed to indicate that the multipolarity of the 688-keV ground-state transition from this state was M2. This would require that the 688-keV state have a spin and parity 2-, in which case direct  $\alpha$  feeding from the 0+ ground state of  $^{240}\text{Pu}$  would be forbidden by parity conservation and therefore unobservable.

This internal contradiction in the experimental results would be removed if the 2- assignment were in error, or if one postulated that  $\alpha$  decay feeds not the 688 keV state, but an additional level which lies slightly above the 688-keV state and decays predominantly to it via a low-energy, unobserved transition. The measured  $\alpha$  energy is consistent with this postulate if the proposed additional level lies not much more than 30 keV above the 688-keV state. However, the  $\alpha$  energy is also consistent with the assumption that the 688-keV state is fed directly. Thus on the basis of these results, the 2- assignment for the 688-keV state and the required corollary postulate of indirect

$\alpha$  feeding were open to doubt. The occurrence of a 2- state at such a low energy in an even nucleus in this region is also suspect on the grounds of systematics; for both reasons it seemed desirable to restudy the decay of this state with high-resolution spectroscopic techniques.

Feeding of the 688-keV state from  $^{240}\text{Pu}$  decay is extremely weak ( $\approx 10^{-5}\%$  of the total  $\alpha$  decays), which makes it very difficult to study the transitions which de-excite it. We therefore studied the decay of  $^{236}\text{Np}$ , which was thought to populate the 688-keV state with an intensity on the order of one percent. These measurements, performed in 1967-1968 with semiconductor spectrometers, have enabled us to confirm the 2- assignment and to establish more thoroughly the properties of the 688-keV state. The multipolarities of the 642- and 688-keV transitions are well established by the K- and L-subshell conversion coefficients, and the state also decays by a weak third transition to the second-excited (4+) state of  $^{236}\text{U}$ . A comparison of the gamma-ray and conversion-electron intensities with those of K-X rays and  $\beta^-$  particles allows us to deduce the absolute branching and log ft for EC decay to the 688-keV level. By measuring the time delay between K-X rays and the 642-keV  $\gamma$  ray, we have also determined the half-life of the level. Together with these results, we include the previously unpublished results of the  $\alpha$ -decay measurements.

## 2. The Alpha Decay of $^{240}\text{Pu}$

Sources of  $^{240}\text{Pu}$  were prepared from available material by vacuum-evaporation from a tungsten filament onto backing foils of either 0.013 cm thick polystyrene or 0.005 cm aluminum. Measurements were performed with a coincidence spectrometer employing a p-n junction silicon detector for  $\alpha$  particles, a NaI(Tl) scintillator for  $\gamma$  rays, and a Si(Li) detector for electrons. Resolution of the fast coincidence circuit,  $2\tau$ , was 40 nanoseconds. Details of the detectors, the fast-slow coincidence circuit, and the methods of energy and intensity calibration are reported elsewhere<sup>1,3</sup>.

Figures 1(a) and 1(b) show spectra of  $\alpha$  particles in coincidence with  $\gamma$  rays and conversion electrons, respectively. Both spectra show the presence of the same  $\alpha$  group in true coincidence. Eight separate measurements of the  $\alpha$  energy yield excitation energies in the range of 673 to 707 keV for the state of  $^{236}\text{U}$  populated by this group. The best value is  $693 \pm 20$  keV, the error being approximately one standard deviation.

Figures 1(c) and 1(d) show the  $\gamma$ -ray and electron spectra in coincidence with the " $\alpha_{693}$ " group. The  $\gamma$  spectrum of a strong  $^{240}\text{Pu}$  source measured with a Ge(Li) detector (fig. 2) shows that the "650-keV  $\gamma$  ray" in fig. 1(c) is actually a doublet consisting of lines at  $642.3 \pm 0.4$  and  $687.8 \pm 0.5$  keV. Partial resolution of the electron lines from these transitions (fig. 1(d)) yields some conversion data which are summarized, together with the rest of the  $\alpha$ -decay data, in Table 1. Although superseded by more precise measurements on  $^{236}\text{Np}$  decay, these data do help to relate the  $\alpha$ -decay data to the EC data, and provide also the  $\alpha$ -transition probability.



As discussed in the introduction, these results suggest the presence in  $^{236}\text{U}$  of a low-lying state of negative parity with  $K = I = 2$ , although it is necessary to postulate that  $\alpha$  decay feeds this state indirectly via a slightly higher-lying level. A plausible assumption is that this level is the spin-3 member of the same band, which should decay to the spin-2 member by a fast ( $\approx 10^{-10}$  s)  $M1+E2$  transition. Our failure to observe direct transitions from the 3- state to the ground-state band implies that the lifetime of the interband transitions is much longer than  $10^{-10}$  s. This inference is confirmed by direct measurement of the half-life of the 688-keV state (see sect. 7).

The 3- member of the  $K\pi = 2-$  band should be about 34 keV above the 2- state, as in  $^{232}\text{U}$  and  $^{234}\text{U}$ . If this state in fact receives the direct  $\alpha$  feeding, the  $\alpha$  energy which we measure is  $\approx 29$  keV too low.

### 3. $^{236}\text{Np}$ Sources

$^{236}\text{Np}$  was produced by the  $^{235}\text{U}(d,n)$  reaction, by bombardment of enriched  $^{235}\text{U}$  with 11 MeV deuterons in the Berkeley 88-inch cyclotron. The low bombarding energy was chosen to minimize the production of  $^{234}\text{Np}$ . Two separate experiments were performed: source I was studied in July 1967, and source II, prepared from a target material of much higher  $^{235}\text{U}$  enrichment, was studied in October 1968. Data on the target material, bombardment conditions, and resulting activities after chemical separation are given in Table 2.

The targets were dissolved in nitric acid and the neptunium coprecipitated on  $\text{LaF}_3$  from a solution 2 M in  $\text{HNO}_3$ , 2 M in  $\text{HF}$ , and 0.16 M in  $\text{H}_2\text{SO}_4$ . An oxidation-reduction cycle eliminated some of the remaining fission products, and a final purification and removal of the carrier La was

then done on an anion-exchange column. The column was washed with many column volumes of 8 M HCl, which removed some, but not all of the remaining Zr and Nb impurities. The activity was then eluted with 2 M HCl and sources were prepared by vacuum sublimation onto thin aluminum backings. The deposits were almost invisible, and the electron lines from the sources showed no noticeable broadening or tailing even at the lowest energies observed (20 keV). The collimators used during the source preparation served as  $\gamma$ -ray sources.

In addition to  $^{236}\text{Np}$ , both sources contained considerable amounts of  $^{238}\text{Np}$ ,  $^{239}\text{Np}$ , and  $^{234}\text{Np}$  produced in the reactions  $^{238}\text{U}(d,n)$ ,  $^{238}\text{U}[(d,n) + (d,p)(\beta^-)]$ , and  $^{235}\text{U}(d,3n)$ . Because the cross sections for the first two of these reactions are both much larger than the  $^{235}\text{U}(d,n)$  cross section, source I contained predominantly  $^{238}\text{Np}$  and  $^{239}\text{Np}$ . The proportion of these isotopes was much lower in source II, due to the much lower proportion of  $^{238}\text{U}$  in the target. In addition, the sources contained on the order of one percent by activity of  $^{97}\text{Zr}$  and its daughter  $^{97}\text{Nb}$ , and very small amounts of  $^{95}\text{Zr}$  and  $^{233}\text{Pa}$ . The presence of other unknown activities was indicated by a few weak, unassigned  $\gamma$  rays.

#### 4. Gamma-Ray Spectra

Spectra of source II were measured with a 13 mm thick  $\times 8 \text{ cm}^2$  planar Ge(Li) detector operated at 4750 volts bias. The resolution of this detector system varies from 1.5 keV at 100 keV to 2.4 keV at 1 MeV, and it exhibits an excellent peak-to-Compton ratio (14:1 for  $^{60}\text{Co}$ ). Source I was studied with a  $35 \text{ cm}^3$  U-drifted coaxial detector which had a resolution of about 4 keV at 1 MeV. Most of the results quoted below are derived from the source II

measurements. Counting circuitry for both the  $\gamma$ -ray and electron detectors has been described elsewhere<sup>5</sup>).

The spectrometers were energy-calibrated with standard sources. The energies of the two prominent  $^{236}\text{Np}$   $\gamma$  rays were also recalibrated by simultaneous measurement with internal standards in order to correct for small drifts. For determination of relative intensities, the photopeak efficiency functions of the detectors were measured with IAEA standard sources.

The half-lives of all but the weakest lines were measured by observing their decay for several weeks following source preparation. These half-lives help to corroborate isotopic assignments based on comparison of energies and energy differences with known level positions in the decay daughters.

In addition to uranium X rays, the  $\gamma$ -ray spectrum contains three lines which we assign to the decay of  $^{236}\text{Np}$ —the previously known transitions and a new weak  $\gamma$  ray at 538 keV. The data are summarized in Table 3, and relevant portions of the spectra are shown in figs. 3(a) and 3(b). Satisfactory agreement between the energy differences and the known energies of transitions within the ground state band of  $^{236}\text{U}$  (Table 3) support the hypothesis that all three lines depopulate the same state at  $687.70 \pm 0.10$  keV. It has also been shown that the 642-keV transition is in coincidence with the 45.3 keV  $2^+ \rightarrow 0^+$  transition, but that the 688-keV transition is not<sup>8</sup>). The present measurements redefine the energy of the  $4^+$  member of this band as  $149.45 \pm 0.15$  keV.

Gamma rays from other activities in the source were examined primarily to ascertain that they should not be assigned to  $^{236}\text{Np}$ . The intensities of  $^{239}\text{Np}$   $\gamma$  rays are in general agreement with published results<sup>9-11</sup>), as are the energies and intensities of  $^{97}\text{Zr-Nb}$   $\gamma$  rays<sup>12</sup>). The energy of the

prominent transition in the decay of  $^{97}\text{Nb}$  is  $658.16 \pm 0.10$  keV. New data obtained on the  $\gamma$  spectrum of  $^{238}\text{Np}$  will be published at a later date. The stronger  $\gamma$  rays of  $^{234}\text{Np}$ , including all previously reported lines above 1 MeV, were observed and their intensities found to be in agreement with values reported by Wapstra<sup>13)</sup> and by Hansen et al.<sup>14)</sup>.

### 5. Conversion Electron and Beta Spectra

Conversion-electron and  $\beta^-$  spectra of the sources were measured with 3 mm thick Si(Li) detectors. The detector used to study source II has a resolution of 2.2 keV at 600 keV. The efficiency of this detector was calibrated relative to the  $\gamma$ -ray detector with sources having  $\gamma$  rays with accurately known conversion coefficients (see fig. 4).

Fig. 5 shows the portion of the spectrum containing lines from the high-energy  $^{236}\text{Np}$  transitions. All conversion lines of the 642- and 688-keV  $\gamma$  rays and the  $L_I(+L_{II})$  line of the 538-keV transition are observed. We also observed the  $L_{II}$  and  $L_{III}$  lines from the low-energy  $2+ \rightarrow 0+$  transitions in  $^{236}\text{U}$  and  $^{236}\text{Pu}$  which follow EC and  $\beta^-$  branching to the  $2+$  levels.

Approximate  $L_I/L_{II}$  ratios for the 642- and 688-keV transitions were obtained by computer-fitting the complex peaks with gaussians with exponential low-energy tails. (Peak positions were fixed with reference to the K-lines and the intensities and shape parameters were allowed to vary.) The resulting fit for the 642-keV transition is shown in fig. 6. Although the presence of the  $L_{II}$  line is not obvious from visual inspection of the figure, the  $L_I+L_{II}$  peak is found from the analysis to be too broad to fit with a single line. The  $L_{III}$  line is well resolved, but its intensity has a large uncertainty due to its

weakness. Similar analysis of the L lines of the 688-keV transitions gives evidence for the existence of a weak  $L_{III}$  line. The errors from these fitting procedures are included in Table 4.

By comparison of the conversion-electron and  $\gamma$ -ray intensities with those from a  $^{137}\text{Cs}$  source and correction for the energy dependence of the detector efficiencies, the absolute conversion coefficients were deduced. These and the subshell-conversion data are summarized in Table 4. Comparison with the theoretical values confirms the M2 assignment for the 688-keV transition. Similar K-conversion data and K/L ratios for the 642- and 688-keV transitions have previously been measured, though not published<sup>8</sup>).

The  $L_{I+II}$  conversion coefficient of the 538-keV transition supports an M2 assignment for this transition also. The 642-keV transition must then be E1 + M2; from the K-conversion coefficient the M2 admixture is  $42 \pm 5\%$ , on the assumption that the E1 and M2 conversion processes are "normal". However, the great retardation of the E1 component, as discussed below, does give rise to the possibility of anomalous E1 conversion. Taken together with the existence of 3  $\gamma$ -ray transitions to  $0^+$ ,  $2^+$ , and  $4^+$  states, the conversion data support an unambiguous  $2^-$  assignment for the 688-keV state.

Although we feel that the transition multipolarities are well established, the measured K/L ratios are about 15-25% lower than the theoretical values<sup>15</sup>). It is difficult to see how this could be due to an experimental effect. A sharp rise in the detector efficiency with increasing energy is both unlikely and inconsistent with the measured efficiency calibration. An alternative possibility, enhancement of the apparent L-line intensity by summing of the K lines with K-X rays, can be discarded because it would have

enhanced predominantly the weak  $L_{III}$  lines, and also because in the same spectrum we observe normal K/L ratios for prominent  $^{239}\text{Np}$  transitions. With due caution regarding the use of an unproven method for measuring precise relative-electron intensities, we feel that the disagreement is very likely a real one. Coupled with the agreement obtained with the theoretical K-conversion coefficient of the 688-keV M2 transition, the low K/L ratios would imply that the  $L_{I+II}$  coefficient is greater than the theoretical value for the 688-keV M2 transition, and probably also for the M2 component of the 642-keV transition. We have no explanation for this discrepancy.

For measurement of  $\beta^-$  continuum spectra, the semiconductor detector has the great disadvantage of an uncertain background which results from Compton distributions of  $\gamma$  rays, and from partial response to electrons due to backscattering. Partially offsetting this disadvantage is the multi-channel nature of the semiconductor spectrometer, because the better statistics thus obtained permit a determination of the endpoint and intensity from a small region near the endpoint.

By extrapolating a linear background from the region beyond the endpoint, (see fig. 5), we obtain a  $\beta^-$  spectrum which yields the Kurie plot shown in fig. 7. The endpoint of the spectrum,  $537 \pm 8$  keV, is somewhat higher than the values previously reported<sup>16,17</sup>). The Kurie plot suggests the presence of a weak inner group. However, the intensity of this group is smaller than the feeding of the 45-keV state of  $^{236}\text{Pu}$ , as deduced from the intensity of the  $L_{III}$  line of the 45-keV transition relative to the intensity of the outer  $\beta^-$  group. Uncertainties in the background probably account for the underestimation of the intensity of the inner group. The data are summarized in Table 5.

## 6. The Decay Scheme of $^{236}\text{Np}$

The measured conversion coefficients relate the intensities of the conversion lines and  $\beta^-$  groups to the intensities of the K-X rays and  $\gamma$  rays. With the use of known fluorescence yields and theoretical electron-capture shell ratios<sup>18</sup>), we have calculated the intensities of primary EC and  $\beta^-$  branches in the decay of  $^{236}\text{Np}$ . The results are given in fig. 8. We have used the ratio (total EC)/(total EC +  $\beta^-$ ) =  $50 \pm 5\%$ , deduced from the ratio of the K-X ray intensity from the decay of  $^{236}\text{Np}$  to the yield of the  $\alpha$ -emitting daughter  $^{236}\text{Pu}$ <sup>17</sup>). This is in good agreement with the value which we derive from the ratio of K-X rays to  $\beta^-$  particles,  $47 \pm 8\%$ .

Log ft values were calculated by using the formulas of Konopinski and Rose<sup>19</sup>). The EC decay energy of  $^{236}\text{Np}$  was deduced to be  $0.96 \pm 0.03$  MeV from the measured  $\beta^-$ -decay energy and closed energy cycles. Most of the uncertainty in this quantity arises from the uncertainty in the  $\beta^-$ -decay energy of  $^{228}\text{Ac}$ .

## 7. The Half Life of the 688-keV Level

The half-life of the 688-keV level was measured by a K-X-ray- $\gamma$ -ray coincidence experiment. The data were taken simultaneously with the  $\gamma$ -ray spectra by splitting the output of the amplifier on the Ge(Li) detector. The 642-keV  $\gamma$  ray was detected by the planar Ge(Li) detector, whose timing characteristics have been carefully studied<sup>20</sup>). K-X rays were detected by a 5-cm  $\times$  5-cm NaI(Tl) scintillator. Leading-edge discriminators on the fast amplifier outputs from each detector produced start-stop signals to a time-to-amplitude converter, the output of which was gated by single-channel windows set at the desired energies.

Response of the system to prompt coincidences was determined with the following sources gated on the energy region indicated:

- a)  $^{22}\text{Na}$             511-keV photopeak [Ge(Li)] - 100-keV Compton [NaI(Tl)]
- b)  $^{60}\text{Co}$             650-keV Compton [Ge(Li)] - 100-keV Compton [NaI(Tl)]
- c)  $(^{234})_{\text{Np}}$         >700 keV [Ge(Li)] - U K-X ray photopeak [NaI(Tl)]

All three sources give approximately the same timing curve. The resolution was 11 ns FWHM, most of which was due to the NaI(Tl) detector. The "prompt" slope on the Ge(Li) side corresponds to a half life of 1.3 ns. The time scale was calibrated by inserting different delays up to 32 ns with a delay box. The uncertainty in this calibration is much smaller than the statistical uncertainties in the delay curve.

Figure 9 shows the 642-keV  $\gamma$ -ray-K-X-ray delay curve with a prompt curve obtained from the  $^{60}\text{Co}$  source for comparison. From the slope on the Ge(Li) side one obtains a half-life of  $4.4 \pm 0.6$  ns for the 688 keV level. The assigned error is larger than the statistical error to allow for possible systematic errors, particularly those which might arise from gain shifts during the 24 hour data-taking period.



## 8. Character of the 2- State

An assignment of 2 for the K-quantum number of the 2- state in  $^{236}\text{U}$  follows from the absence of any lower-lying member of the band. Such a level would certainly have been populated by a rotational transition within the lifetime of the 2- state. Although the reduced branching ratios (Table 6) are closer to those expected for  $K = 2$  than for  $K = 0$  or 1, the agreement is poor. It is possible that our estimate of the M2 component of the  $2- \rightarrow 2+$  transition is high if the E1 component is anomalously converted. The high intensity for branching to the 4+ state could be accounted for by a small  $K = 1$  or  $K = 0$  impurity, but the assumption of such an impurity would lead to serious difficulties in accounting for the large retardation of the E1 transition rates, as discussed below. This discrepancy, as well as the high  $(2 \rightarrow 2)/(2 \rightarrow 0)$  branching ratio, may indicate the need for higher-order corrections to the Alaga rules<sup>21</sup>).

The 2- state of  $^{236}\text{U}$  is unusual in several respects. Its energy is low—in fact, it is the lowest known 2- state in any even nucleus. It is also characterized by relatively fast M2 transitions and greatly retarded E1 transitions to the ground-state band. Although little is known about other 2- states in the deformed region, there are well characterized 2- states in  $^{232}\text{U}$  and  $^{234}\text{U}$  (22-24), some properties of which are compared in Table 7. A trend toward lower energy, slower E1's, and faster M2's with increasing neutron number is evident. There is presently no experimental information on 2- states in Th or Pu which might yield trends with respect to proton number. The hindrance factors for  $\alpha$  decay to the spin-3 members of the bands are comparable to those of the spin-1 members of the  $K = 0$  octupole bands in this

region<sup>1</sup>), but are lower than the hindrance factors for spin-3 members of the same  $K = 0$  bands<sup>1,28</sup>).

Calculated energies of  $K = 2$  octupole states increase with increasing neutron number<sup>26,27</sup>). As pointed out by Faessler and Plastino<sup>26</sup>), the calculated energies depend mainly on the well-established positions of single-particle orbitals rather than on the specific form of the potential interaction. Thus the calculations fail to reproduce the trend in the energies. The same calculations do reproduce the trend toward lower 2- states in Cm and Cf isotopes.

The calculations of Faessler and Plastino and of Donner and Greiner<sup>29</sup>) also predict hindrance factors on the order of 100 for E1 transitions from the  $K = 1$  and  $K = 0$  bands. Since the hindrance factor for the  $2- \xrightarrow{E1} 2+$  transition in  $^{236}\text{U}$  is  $2 \times 10^7$ , either the admixture of  $K = 0$  or  $K = 1$  in the 2- state is  $< 10^{-5}$ , which implies a surprising degree of K-purity, or there is a fortuitous cancellation of  $K = 0$  and  $K = 1$  contributions to the E1 matrix elements.

If one includes the statistical factor  $\left[ \begin{matrix} I_i & l & I_f \\ C_{K_i} & & K_f - K_i \end{matrix} \right]^2$  in the theoretical estimate, the M2 transitions in  $^{236}\text{U}$  are close to the single-particle estimate. Such fast transitions must arise from major components of the wave functions of the initial and final states. There is presently no direct information on the structure of the 2- state in  $^{236}\text{U}$ . The major component of the 2- state in  $^{234}\text{U}$  is the two-neutron configuration  $7/2-[743] 3/2+[631]$ ; from the spectroscopic factor for this state in the reaction  $^{235}\text{U}(d,t)$ , Bjørnholm et al.<sup>24</sup>) deduced that the squared amplitude of this component is  $0.58 \pm 0.10$ . The calculations of Soloviev and Siklos<sup>27</sup>) predict a value near 0.75 for the 2- states of both  $^{234}\text{U}$  and  $^{232}\text{U}$ , with an additional component ( $\approx 15\%$ ) of the

two-proton configuration  $1/2-[530] 5/2+[642]$ . These two components cannot by themselves account for the strength of the M2 transitions in  $^{236}\text{U}$ . Transitions between the bands  $1/2-[530]$  and  $5/2+[642]$  in  $^{237}\text{Np}$  are hindered by a factor of  $10^{2-30}$ ). The transition  $7/2-[743] \xrightarrow{\text{M2}} 3/2+[631]$  has not been observed, but if it were hindered by less than a factor of  $10^2$ , it should have resulted in observable M2 transitions in the decay of the 2- state of  $^{234}\text{U}$ .

In the absence of any other probable strong component of the 2- state which can be constructed from two Nilsson orbitals connected by allowed<sup>31)</sup> M2 transitions, it is interesting to speculate that the M2 transitions in  $^{236}\text{U}$  may result from a coherence effect. Such a "collective" M2 could be related to the apparent collectivity of the state, as suggested by its low energy. It would be especially interesting to Coulomb-excite the 3- member of the band, in order to determine whether the B(E3) also exhibits a high value characteristic of octupole-vibrational states.

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TABLE 1  
Weak  $\alpha$  branches of  $^{240}\text{Pu}$

	$\alpha$ Groups	
Alpha energy, MeV	4.48	4.21 ?
Excitation energy, keV	693 $\pm$ 20	970 $\pm$ 50
Intensity, coinc. with $\gamma$ rays	$(1.8\pm 0.3)\times 10^{-5}\%$	$<1\times 10^{-6}\%$ <sup>a)</sup>
Intensity, coinc. with electrons	$(3.4\pm 1.0)\times 10^{-6}\%$	$\leq 3\times 10^{-7}\%$
Total intensity	$(2.1\pm 0.4)\times 10^{-5}\%$	$<1.3\times 10^{-6}\%$
Hindrance factor <sup>b)</sup>	55	>8
<u><math>\gamma</math> rays coincident with <math>\alpha_{693}</math></u>		
Gamma energy	642.3 $\pm$ 0.4 <sup>c)</sup>	687.8 $\pm$ 0.5 <sup>c)</sup>
Relative intensity	1.00 <sup>c)</sup>	0.27 $\pm$ 0.02 <sup>c)</sup>
$\gamma$ rays/ $^{240}\text{Pu}$ $\alpha$ decay	$(1.4\pm 0.2)\times 10^{-5}\%$	$(0.38\pm 0.10)\times 10^{-5}\%$
$e_{\text{K}}/\gamma$	0.11 $\pm$ 0.04	0.26 $\pm$ 0.10
K/L+M+...	3.5 $\pm$ 1.0	3.0 $\pm$ 1.2

<sup>a)</sup>We obtain scant evidence (5 counts) for a  $\gamma$  ray of  $\approx 860$  keV in coincidence with this  $\alpha$  group.

<sup>b)</sup>Hindrance factors are calculated with the one-body equations of Preston <sup>4)</sup>; the nuclear radius was chosen to make  $\text{HF} = 1$  for the ground state transition.

<sup>c)</sup>From the  $\gamma$ -ray singles spectrum.

TABLE 2  
Sources for  $^{236}\text{Np}$  studies

	<u>Source I</u>	<u>Source II</u>
Target material:	uranium foils	$\text{U}_3\text{O}_8$ packed powder
Isotopic composition:		
$^{234}$		0.007%
$^{235}$	93%	99.71 %
$^{236}$		0.022%
$^{238}$	7%	0.26 %
Bombardment:		
energy at target	11 MeV	11 MeV
flux	10 $\mu\text{A}$	20 $\mu\text{A}$
length of bombardment	3.3 hours	6 hours
Composition of source (by activity): a)		
$^{234}\text{Np}$	1.5	not measured
$^{236}\text{Np}$	<u>100</u>	<u>100</u>
$^{238}\text{Np}$	56	3.1
$^{239}\text{Np}$	124	8.5
$^{233}\text{Pa}$	not observed	0.004
$^{97}\text{Zr-Nb}$	0.8	1.9
$^{95}\text{Zr-Nb}$	not observed	0.17

a) Intensities relative to the  $^{236}\text{Np}$  disintegration rate at the end of the bombardment.



TABLE 3  
 $\gamma$  rays of  $^{236}\text{Np}$

$E_{\gamma}$ , keV	Relative intensity	Half-life (hours)
U K $_{\alpha_2}$	1047±50	22.5±0.4
U K $_{\alpha_1}$	1712±85	
U K $_{\beta_1}$	599±50	
U K $_{\beta_2}$	not well resolved	
538.25±0.20	1.11±0.12	23±6
642.42±0.10	<u>100</u>	22.4±0.6
687.71±0.10	26.5±0.5	22±2

Energy differences

Measured difference	Ground-state band transition
687.71-642.42 = 45.29±0.07	45.28±0.06 <sup>6)</sup>
642.42-538.25 = 104.17±0.15	103.6 ±0.3 <sup>7)</sup>

TABLE 4  
Conversion coefficients of  $^{236}\text{Np}$   $\gamma$  rays

Coefficient and transition	Experimental value	Theoretical <sup>a)</sup>					
		E1	E2	E3	M1	M2	
$e_K/\gamma$	642	0.112±0.010	0.00950	0.0186	0.0451	0.109	0.251
	688	0.22 ±0.02	0.00598	0.0165	0.0394	0.0908	0.207
$e_{L_{I+II}}/\gamma$	538	0.086±0.027	0.00156	0.00975	0.0532	0.0330	0.101
K/L	642	3.59 ±0.11	5.73	2.79	1.48	5.29	4.19
	688	3.27 ±0.16	5.75	2.99	1.66	5.26	4.26
$L_I/L_{II}$	642	11±4	6.2	1.24	0.61	8.1	6.3
	688	7±3	6.5	1.40	0.69	8.2	6.3
$L_{I+II}/L_{III}$	642	$36^{+10}_{-7}$	13.9	9.4	10.3	254	43
	688	$46^{+40}_{-20}$	14.5	10.5	11.4	256	48

<sup>a)</sup> Interpolated from the tables of Hager and Seltzer<sup>15)</sup>.

TABLE 5  
Beta groups of  $^{236}\text{Np}$

	Present work	Gray <sup>17)</sup>	O'Kelly <sup>16)</sup>
$E_0(1)$ , keV	537±8	500±30	518±10
$E_0(2)$ , keV	[492] <sup>a)</sup>		
$I(2)/I(1)$	0.07 <sup>b)</sup>		
$I_{LM...}(44.6 \gamma)/I_{\beta}$ <sup>c)</sup>	0.25±0.09 <sup>b)</sup>	0.17	
$I_{LM...}(45.3 \gamma)/I_{\beta}$ <sup>c)</sup>	0.16±0.06	0.17	
$I_K(642 \gamma)/I_{\beta}$	$(2.0\pm0.6)\times 10^{-3}$		

<sup>a)</sup>The Kurie plot was fit with two components with the constraint that the difference between their endpoints be 45 keV.

<sup>b)</sup>According to the decay scheme these numbers should be equal. The discrepancy probably indicates an underestimation of the intensity of the inner beta group. We have adopted the intensity of the 44.6-keV transitions as the beta feeding of the 2+ level of  $^{236}\text{Pu}$ .

<sup>c)</sup>The intensities of the low-energy transitions are calculated from the intensities of the  $L_{III}$  lines and the theoretical subshell ratios.

TABLE 6

M2 branching ratios in the decay of the 2- state

Transition	Reduced branching ratio <sup>a)</sup>			
	Observed	K = 0	K = 1	K = 2
538 (2- → 4+)	14±2	250	115	7.5
642 (2- → 2+)	224±27 <sup>b)</sup>	143	36	143
688 (2- → 0+)	(100)	(100)	(100)	(100)

<sup>a)</sup>Theoretical estimates for K = 0, 1, or 2 are based on the Alaga rules<sup>21)</sup>.

<sup>b)</sup>On the assumption that the E1-component of this transition is not anomalously converted.

TABLE 7

Some properties of the  $K = 2$  negative parity bands in uranium isotopes

	$^{232}\text{U}$	$^{234}\text{U}$	$^{236}\text{U}$
E, keV	observed	1017	688
	calculated, ref. 26	1130	1270
	calculated, ref. 27	1100	-----
$t_{1/2}$ , s	$<5 \times 10^{-11}$ <sup>22)</sup>	$\approx 2 \times 10^{-10}$ <sup>b)</sup>	$4.4 \times 10^{-9}$
F(M2, 22- $\rightarrow$ 02+) <sup>a)</sup>	-----	$>100$ <sup>c)</sup>	8
F(E1, 22- $\rightarrow$ 02+) <sup>a)</sup>	$<4 \times 10^5$	$\approx 2 \times 10^6$	$2 \times 10^7$
HF ( $\alpha$ , 00+ $\rightarrow$ 23-)	-----	$\approx 21$ <sup>28)</sup>	55

<sup>a)</sup>  $\gamma$ -ray hindrance factors are relative to the Moszkowski estimates for a single proton transition <sup>25)</sup>; the statistical factor is omitted from the theoretical estimate.

<sup>b)</sup> Estimated from the competition between interband E1 and intraband transitions <sup>24)</sup>.

<sup>c)</sup> From the agreement of the K-electron line intensity with the  $\gamma$ -ray intensity and the theoretical E1 conversion coefficients <sup>23,24)</sup>, we estimate that the M2 component of this transition is  $\leq 1\%$  in both  $^{234}\text{U}$  and  $^{232}\text{U}$ .

## Figure Captions

Fig. 1. Coincidence spectra of  $^{240}\text{Pu}$

(a)  $\alpha$  particles coincident with  $\gamma$  rays of energy  $>500$  keV.

(b)  $\alpha$  particles coincident with electrons of energy  $>450$  keV.

(c)  $\gamma$  rays coincident with the  $\alpha_{693}$  group. The source used in this measurement contained  $\approx 2\%$   $^{239}\text{Pu}$  by activity. Sources used for other coincidence measurements contained  $<0.2\%$   $^{239}\text{Pu}$ .

(d) Conversion electrons coincident with  $\alpha_{693}$ .

Fig. 2.  $\gamma$  rays of a strong  $^{240}\text{Pu}$  source. The source contained  $0.7\%$   $^{241}\text{Am}$  by activity.

Fig. 3.  $\gamma$ -ray spectrum of  $^{236}\text{Np}$

(a) K-X-ray region. The Pu X-rays are due to  $^{238}\text{Np}$  and  $^{239}\text{Np}$ .

(b) Region of the  $^{236}\text{Np}$   $\gamma$  rays.

Fig. 4. Efficiency of the Si(Li) detector. The standard sources were  $^{109}\text{Cd}$ ,  $^{113}\text{Sn}$ ,  $^{207}\text{Bi}$ ,  $^{137}\text{Cs}$ , and  $^{239}\text{Np}$ .

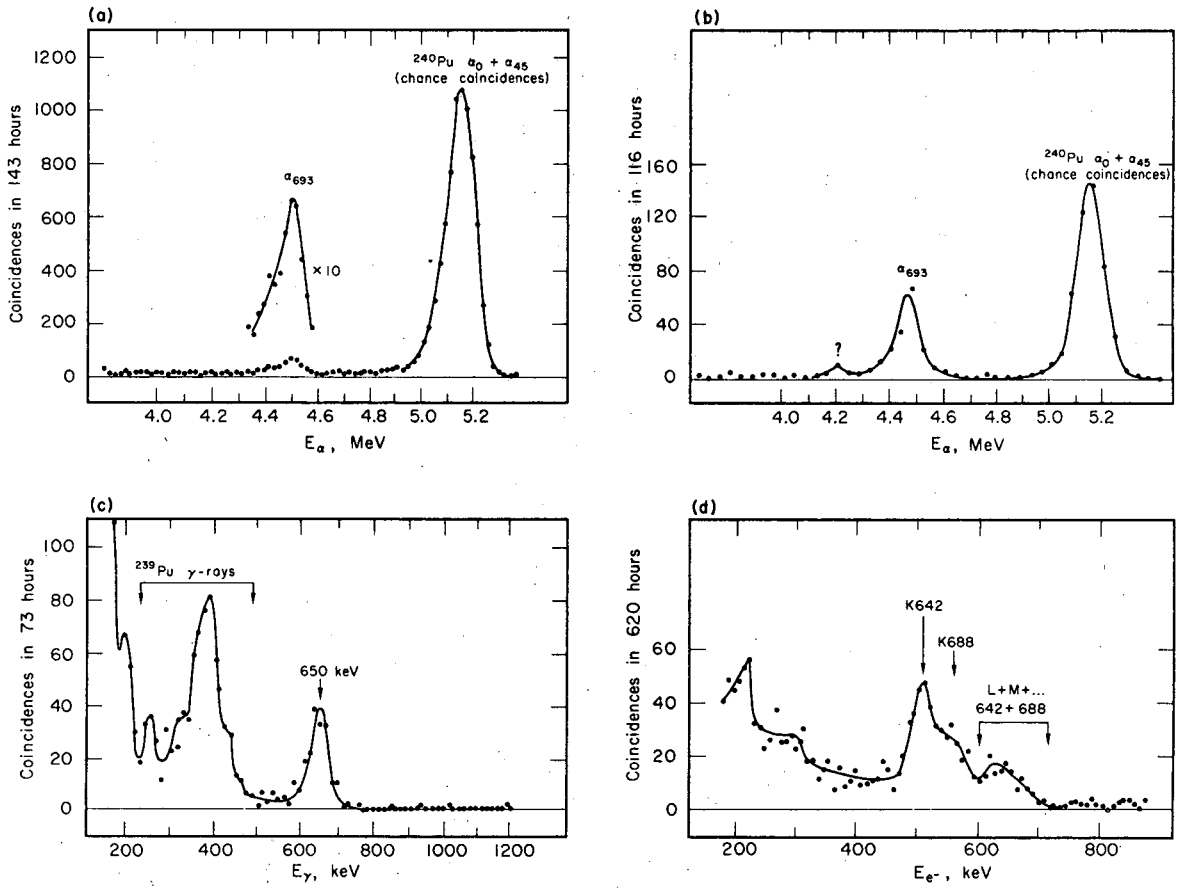
Fig. 5. A portion of the  $^{236}\text{Np}$  electron spectrum. The background extrapolation is that used for analysis of the  $\beta^-$  continuum spectrum.

Fig. 6. Computer fit of the L lines of the 642-keV transition. The peak positions were fixed relative to the K line, and the intensities, shape parameters, and the linear background were varied to produce a least-squares fit.

Fig. 7. Kurie plot of the  $^{236}\text{Np}$  beta spectrum near the endpoint.  $F'$  is the Fermi function, corrected for screening<sup>19</sup>).

Fig. 8. Decay scheme of  $^{236}\text{Np}$ . The postulated indirect  $\alpha$  feeding of the 688-keV state of  $^{236}\text{U}$  is also shown. Slanted numbers represent  $\log ft$  values and  $\alpha$ -hindrance factors.

Fig. 9. Delay curve for K-X-ray-642-keV  $\gamma$ -ray coincidences.



XBL6812-7492

Fig. 1 (a-d).



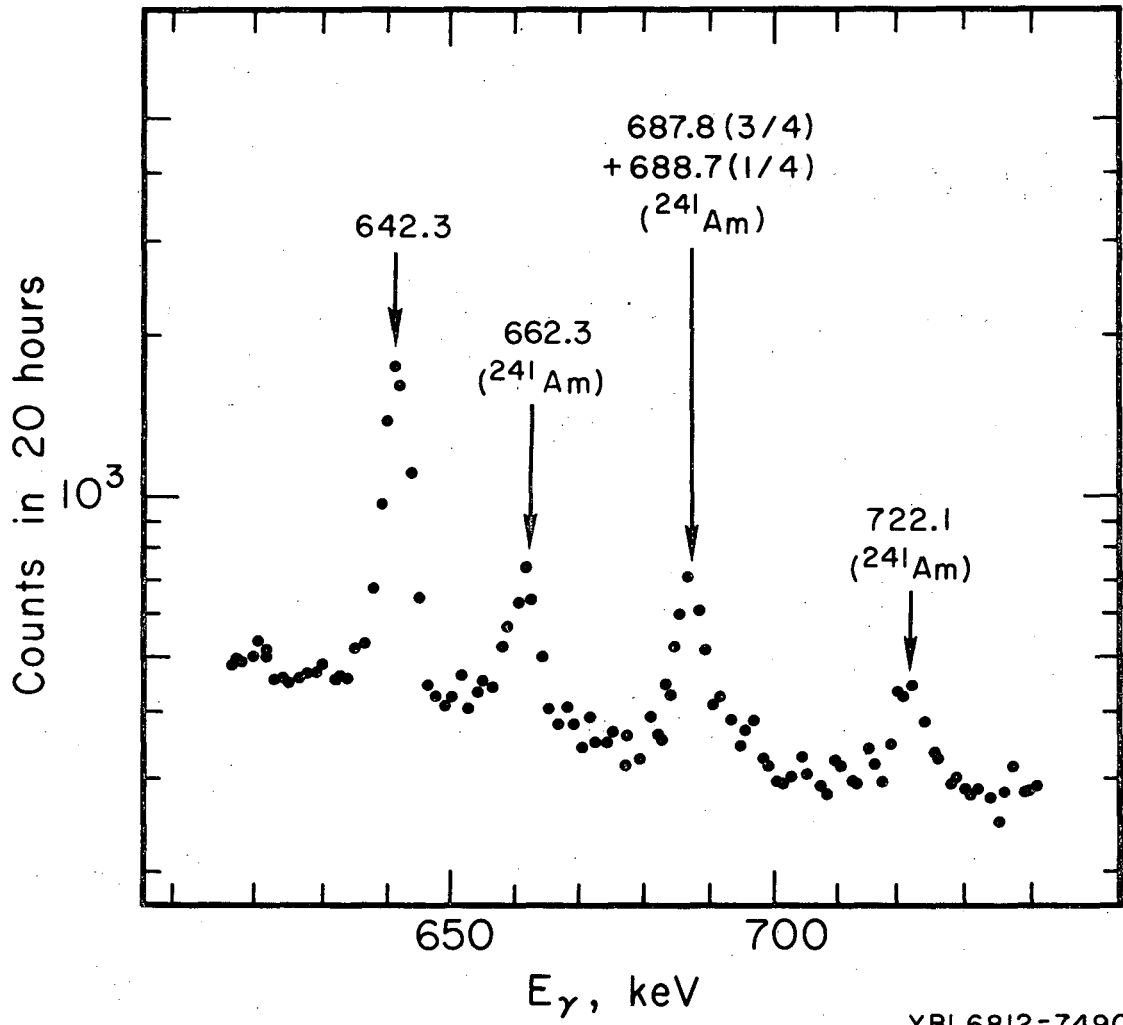


Fig. 2.

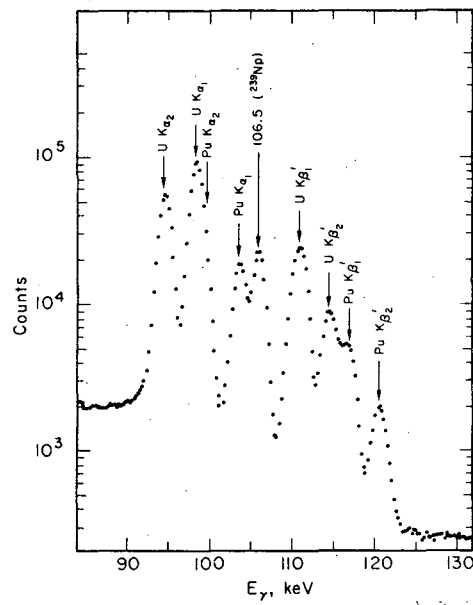


Fig. 3 (a).

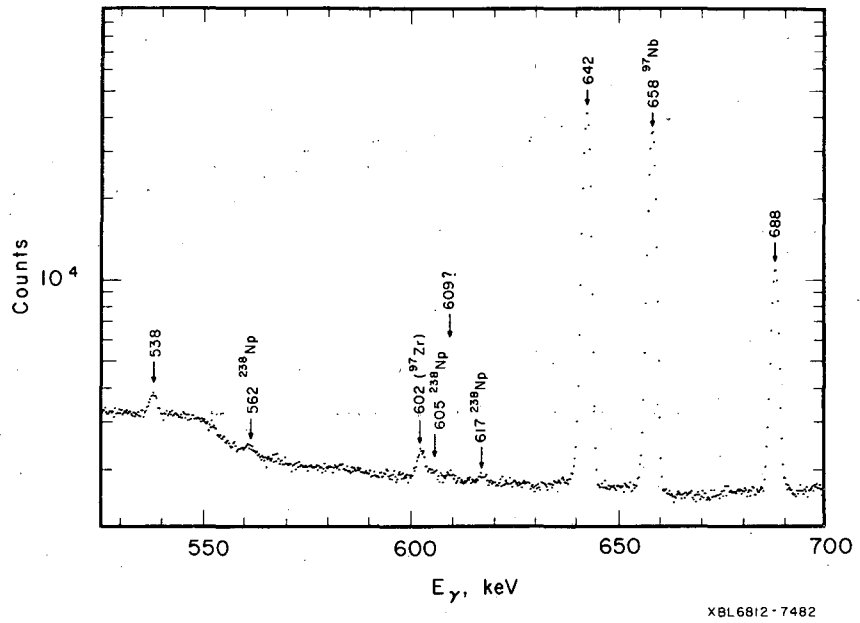
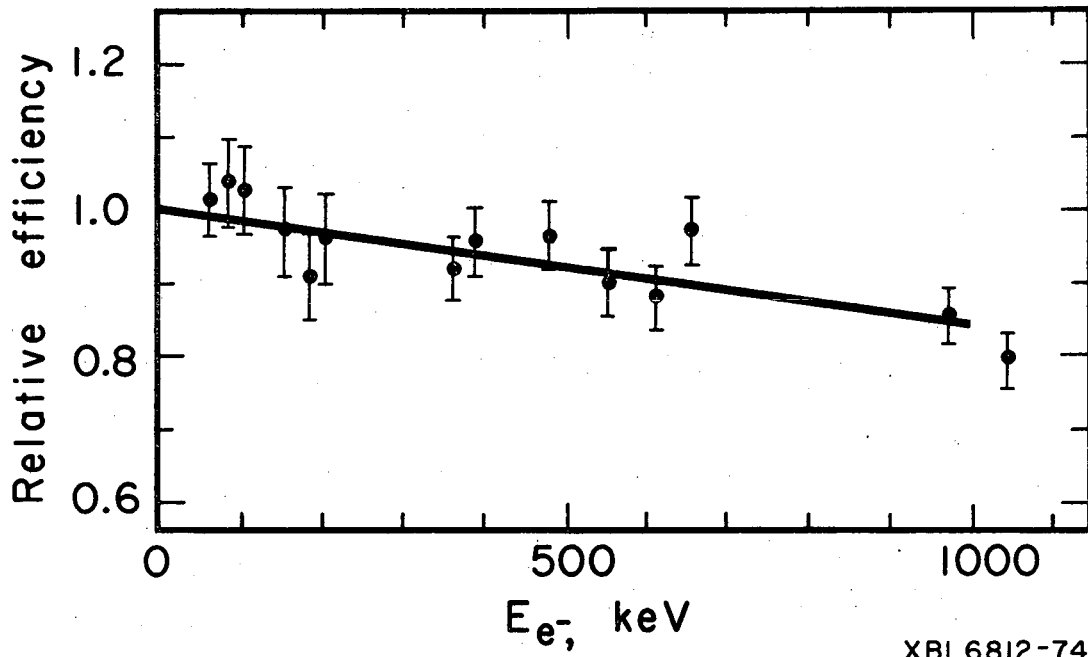


Fig. 3 (b).



XBL6812-7489

Fig. 4.

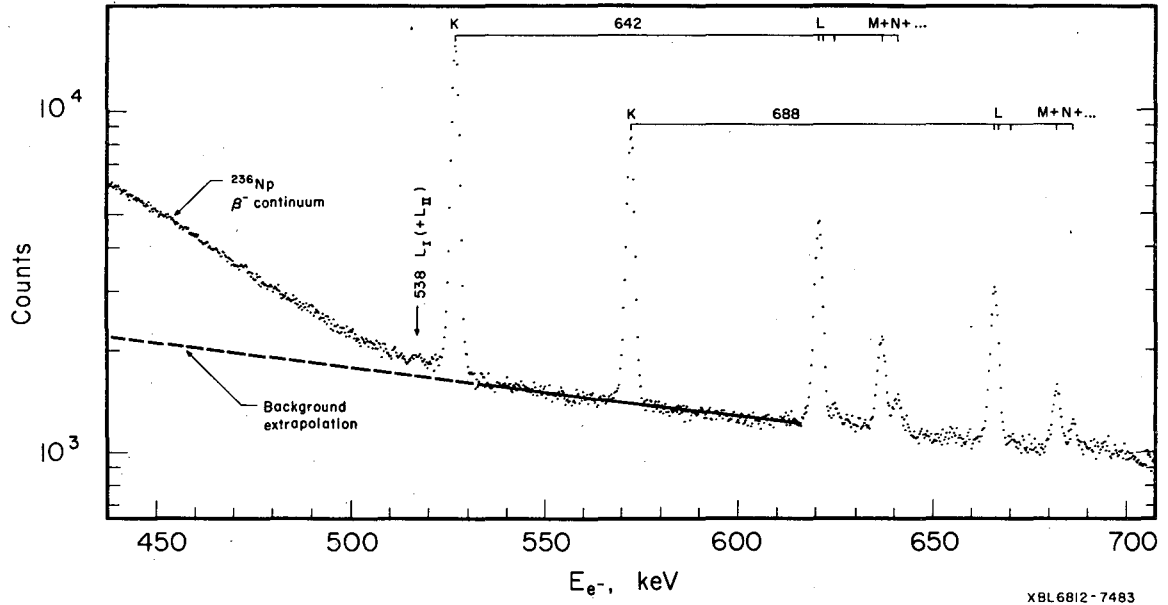
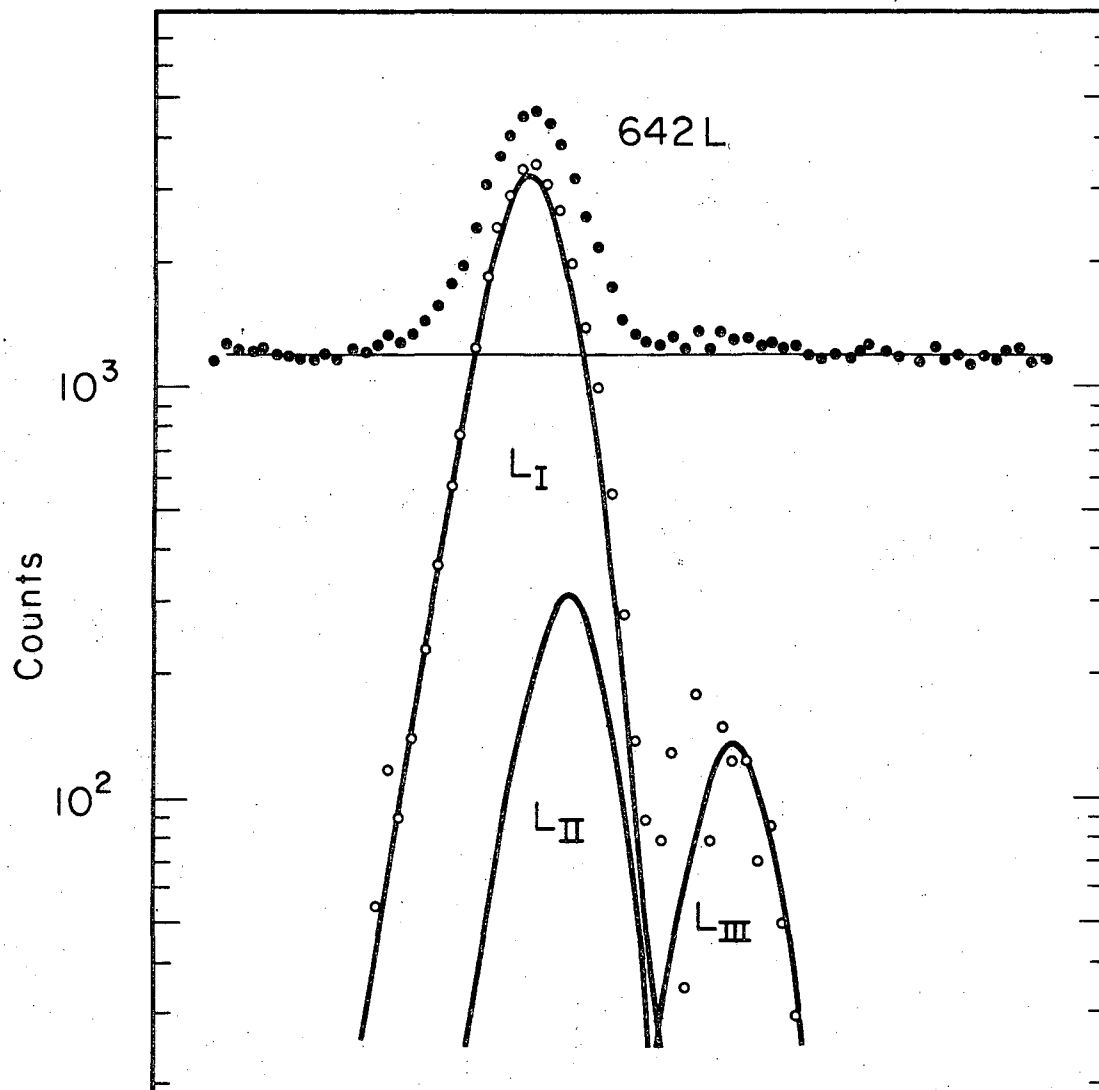
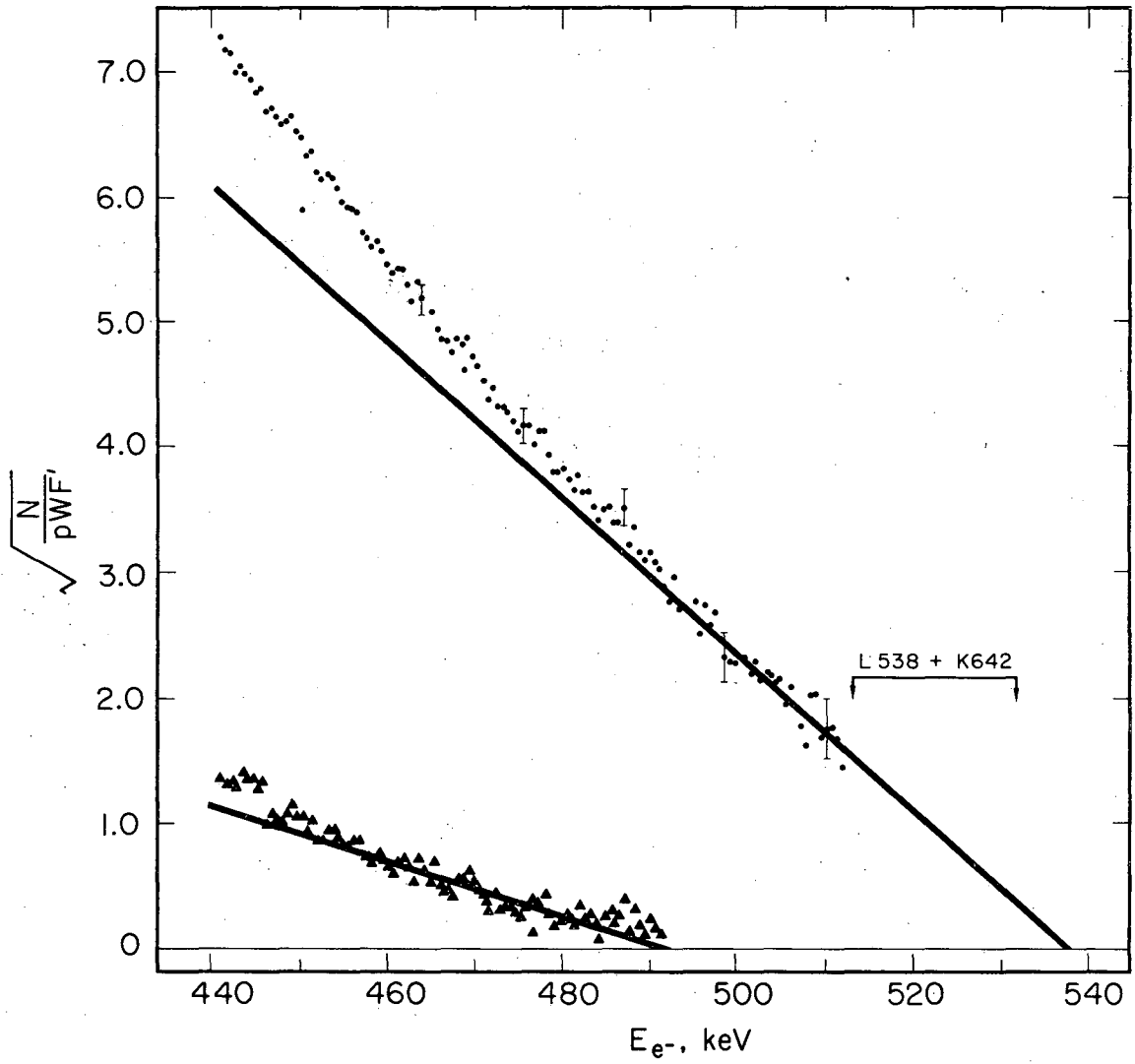


Fig. 5.



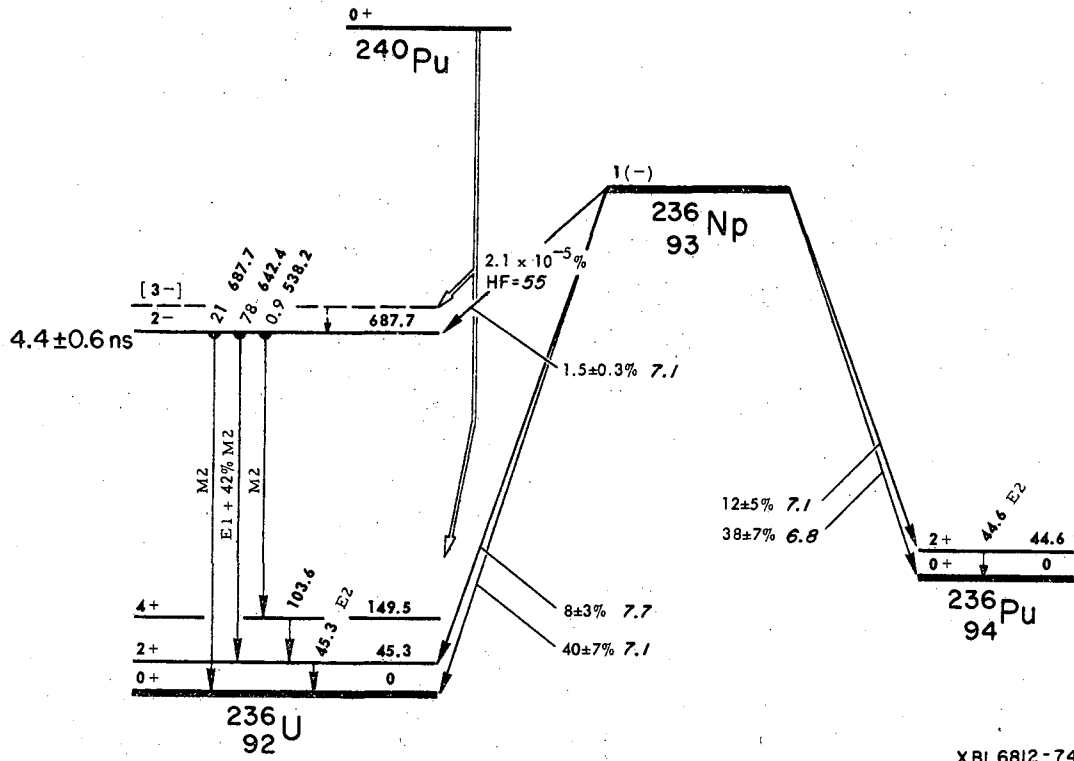
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Fig. 6.



XBL6812-7495

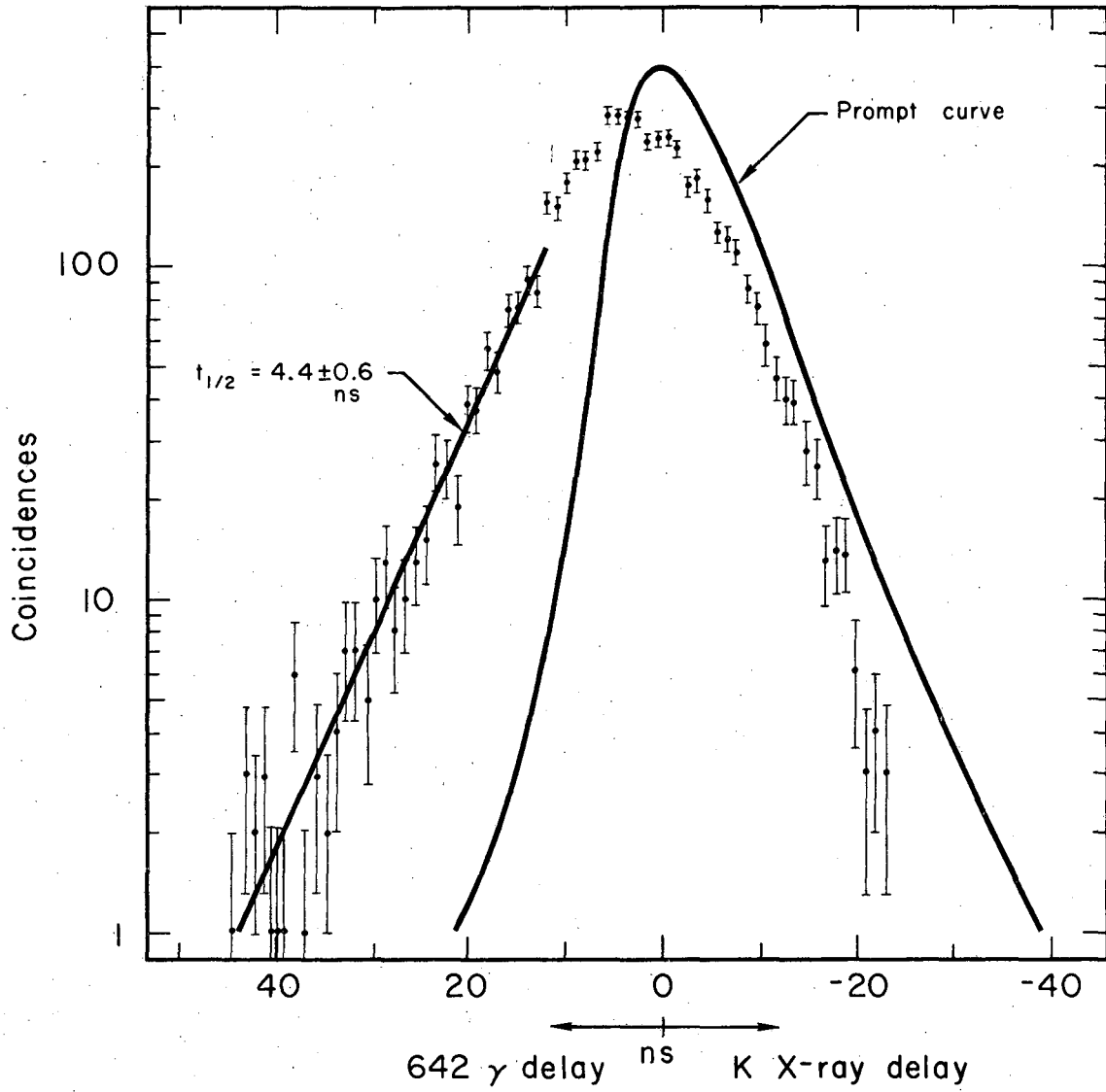
Fig. 7.



XBL6812-7488

Fig. 8.





XBL6812-7487

Fig. 9.

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