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ENERGY LEVELS IN ^{235}Np*

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ABSTRACT

The energy levels in ^{235}Np have been studied through the alpha decay of ^{239}Am and the electron capture decay of ^{235}Pu. A level was observed at 34.1 keV and assigned to the ground state rotational band with Nilsson quantum numbers 5/2 + [642]. Levels at 49.0, 91.6, and 146.8 keV were assigned to the 5/2 - [523] rotational band. The lifetime of the 49.0 keV state was measured as 6.9 nanoseconds, and the half-life of the ^{235}Pu was redetermined to be 24.3 min.

INTRODUCTION

A considerable amount of work has been done in the determination of the energy levels in ^{237}Np and ^{239}Np,[1,2] however no comparable body of data exists for ^{235}Np.

The energy levels of ^{235}Np are populated in the alpha decay of ^{239}Am and in the electron capture decay of ^{235}Pu. Previous work[3,4] has shown that ^{239}Am decays to a 48-keV state in ^{235}Np by emission of a 5.78 MeV alpha particle and that this state de-excites to the ground state by an E1 transition.

We have looked at the alpha spectrum of ^{239}Am with a resolution of 18 keV full width at half maximum (FWHM) using a surface barrier Au-Si detector and have observed four alpha groups.

^{235}Pu decays by electron capture (>99%) and alpha emission (.003%).[5,6] We have studied its decay using high resolution (1 - 2 keV FWHM) Ge(Li) detectors in singles and coincidence measurements. In the gamma singles spectrum we observed copious K and L X-rays and two low energy gamma rays of 34.1 and 49.0 keV. The lifetime of the 49 keV state was measured and the half-life of the ^{235}Pu was redetermined.

SOURCE PREPARATION

Americium-239 was prepared in a (d, 2n) reaction by bombarding ~ 300 mg of ^{239}Pu fluoride with 18 MeV deuterons in the 88 inch cyclotron at the Lawrence Radiation Laboratory, Berkeley. The americium was separated from the plutonium by passing the sample through an AG 1-X8 anion column in 10M HCl.[7] After adding lanthanum carrier, the americium was taken through a $La(OH)_3$ - LaF_3 cycle.[8] The lanthanum carrier was separated from the americium on an AG 50-X12 cation column with a 20% ethanol-saturated HCl

solution.[8] After a final purification on an AG 50 cation cleanup column,[9] the americium was vaporized onto platinum. Due to the presence of ^{241}Pu in the source material all of the sources prepared contained ^{241}Am.

The ^{235}Pu was prepared in an (α, 4n) reaction by bombarding ^{235}U metal foil with 45 MeV alpha particles in the 88 inch cyclotron. The plutonium was separated from the uranium by elution from an AG 1-X8 anion column with 10M HCl - 0.1M HI solution.[7] The plutonium was further purified by adding lanthanum carrier and taking it through a $La(OH)_3$ - LaF_3 cycle.[8] The lanthanum was removed on a AG 1-X8 anion column by elution with 10M HCl, and the solution containing the plutonium was evaporated to dryness on a mylar film. The chemistry was completed in 2-3 hours.

^{239}Am ALPHA DECAY

The ^{239}Am alpha spectrum obtained on the solid-state detector is shown in Fig. 1. The spectrum has been smoothed by a computer[10] to make the smaller peaks stand out more clearly above the background. It can be seen that the spectrum is almost identical to that of ^{241}Am. The peak at 5.38 MeV in the spectrum has been resolved into two alpha groups, one of which is the α_{159} of ^{241}Am. The other is the first observation of alpha decay in ^{240}Am and will be the subject of another publication.[24]

Table I lists the ^{239}Am alpha groups, with their intensities and calculated hindrance factors.

The theoretical alpha intensities for favored alpha decay can be calculated using the equation[21,22]

$$P = P_o(Z,E) \sum_L C_L \left| < I_i \; L \; K_i \; K_f - K_i \, | I_f \; K_f > \right|^2$$

$P_o(Z,E)$ is the transition probability calculated from simple spin-independent alpha-decay theory,[20] the subscripts i and f refer to the initial and final states respectively, and the term in brackets is the Clebsch-Gordan coefficient. The parameter C_L is determined empirically from known alpha intensities and is inversely proportional to the hindrance factor of the alpha wave of angular momentum L.

The results of such a calculation are summarized in Table II, where it is seen that our measured alpha intensities agree quite well with those predicted for favored alpha decay.

The hindrance factor for the ground state alpha decay of ^{239}Am (see Table I) is large (1300); similar to both ^{241}Am (1100) [5] and ^{243} (1900).[5] This highly hindered alpha decay is characteristic of decay between states with different Nilsson quantum numbers.[25]

^{235}Pu ELECTRON CAPTURE DECAY

In the gamma singles spectrum of ^{235}Pu we observed the K and L X-rays of Np, and two gamma rays of energy 49.0±0.1 and 34.1±0.1 keV. The intensities of these gamma rays were determined in both singles and coincidence measurements and these are given in Table III. The decay was followed for several hours and the half-life was determined to be 24.3±0.1 min from measurements on the K X-rays.

THE HALF-LIFE OF THE 49 keV STATE

The lifetime of the 49 keV state was determined by the delayed coincidence method using a ^{235}Pu source. Two 3" × 3" NaI(Tl) crystals were

used; one was gated on the K capture X-rays, and the other on the 49 keV gamma ray. The two outputs were fed into a time-to-pulse-height converter and the time delay spectrum recorded in a 400 channel pulse height analyzer. A prompt spectrum was obtained using a source of ^{182}Ta and performing the coincidence between the compton distribution of the 229 and 100 keV gamma rays using the same low energy gate (49 keV) as for the ^{235}Pu.

The simplest method of determining the lifetime of a level in a delayed coincidence experiment is to measure the slope of the delayed curve. However, this is not possible when the half-life of the radiation is short compared to the time resolution of the equipment, as it is in this case. The data were therefore subjected to a least squares analysis in which the delayed curve is constructed from the prompt curve by introducing a delay of mean life $\tau = 1/\lambda$. The sum of the squares of the residuals between the measured and calculated delayed curves is minimized with respect to λ.[11] The result of the least squares analysis is shown by the solid curve in Fig. 2. The best fit corresponds to a half-life for the 49 keV state of 6.9 nanoseconds. Several other methods of analysis were also used and they are summarized in Table IV.

The best value of the half-life of the 49 keV level obtained as a weighted average of the different methods is 6.9±0.3 nanoseconds.

ANALYSIS OF DATA

The energy levels of ^{235}Np are very similar to those of ^{237}Np and ^{239}Np. This is clearly seen in Fig. 3.

From the alpha and gamma spectra we place energy levels in ^{235}Np at 34.1±0.1, 49.0±0.1, 91.6±0.3, 146.8±0.7 keV. By analogy with the decay of ^{241}Am and ^{243}Am, the levels at 49.0, 91.6, and 146.8 keV likely belong to the same rotational band. If we take the measured energy differences and substitute

into the simple rotational formula to solve for the spin of the band head I_o, we get

$$\frac{E(I_o + 1, I_o + 2)}{E(I_o, I_o + 1)} = \frac{I_o + 2}{I_o + 1} = 1.30 \pm 0.02$$

$$I_o = 2.38 \pm 0.17$$

which is 5/2 within the experimental error.

Fitting to the 49.0 and 91.6 keV levels we obtain a rotational constant of 6.09±0.04 keV. Using this we would predict the 9/2 member to fall at 146.4±0.5 keV, which fits the 146.8 keV level within the experimental error.

In a coincidence experiment with the unresolved alpha spectrum we observed a gamma ray of 49 keV energy. Earlier work by Asaro et al.[4] has shown this to be an E1 transition. The ground state of ^{235}Np has previously been assigned the Nilsson quantum numbers 5/2 + [642].[16,17,18] This is consistent with the assignment of spin 5/2 and negative parity to the 49 keV level. It very likely has the Nilsson assignment 5/2 - [523] as do analogous levels in ^{237}Np and ^{239}Np.

Fig. 4 shows our proposed decay scheme.

The intensity of the 49 keV gamma ray in coincidence with K X-rays is 1.7%. It had been previously determined from ^{239}Am decay[4] that this gamma ray was E1 with an intensity of 0.5 per alpha disintegration. The theoretical conversion coefficient for an E1 transition of that energy is 1. Therefore about 4% of the K capture populates the 49 keV state directly or via higher states. Approximately 96% of the K capture processes then bypass this state and very likely populate the ground state rotational band.

The energy levels of ^{235}Np are similar to those of ^{237}Np and ^{239}Np.[1,5] The first excited state in each of these latter two nuclides lies at about 32 keV and decays to the ground state by predominantly M1 radiation with some E2 admixture. If we assume the same admixture as for ^{237}Np (1.4% E2) [5] the theoretical conversion coefficient for the 34.1 keV gamma ray would be 195. The intensity of the 34.1 keV gamma ray in coincidence with K X-rays is 0.2%. Therefore the intensity of the transition is estimated to be about 40%. This intensity is sufficiently large that it cannot come from de-excitation of the 49 keV state, and hence it very likely represents K capture to the 34.1 keV level. The remaining ~ 56% of the K capture very likely populates the ground state.

L_1/K CAPTURE RATIO

Using the equations of Brysk and Rose[5,19] it is possible to calculate the expected L_1/K capture ratio to the 49 keV state. Assuming that it is a first forbidden non unique transition, and taking the Q-value for electron capture calculated from closed decay energy cycles as 1.13 MeV, the calculated L_1/K ratio is 0.21.

Using the experimental data, the difference between the singles and coincidence measurements represents the number of gamma rays that are not in coindidence with K X-rays, and hence the number that are in coincidence with L, M, N, etc. X-rays. Therefore taking EC(M + N + ...)/EC(L) = 0.52, $EC(L_{11})/EC(L_1) = 0.14$, and $EC(L_{111}) = 0$,[5] our measured L_1/K ratio is 0.31±0.10. Although this fits the calculated value within the experimental error, it is not sufficiently precise to be of significant use in determining the Q-value.

The K capture process in ^{235}Pu populates the 5/2 and 7/2 members of the ground state rotational band, indicating that ^{235}Pu should have spin 5/2 or 7/2. This spin would be consistent with the K capture population to the 5/2 - [523] band also. By analogy to ^{233}U which has the same number of neutrons, the ^{235}Pu ground state probably has the Nilsson assignment 5/2 + [633].

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FOOTNOTES AND REFERENCES

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†National Research Council of Canada Postdoctoral Fellow. 1968-1969.

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Table I. ^{239}Am Alpha Groups

Alpha Particle Energy (MeV)	Excited State Energy (keV)	Intensity (%)	Hindrance Factor
5.825±0.004	0	0.33±0.02	1300
5.776±0.002	49.0±0.1	83.7 ±0.4	2.9
5.734±0.002	91.6±0.3	13.75±0.07	10
5.680±0.002	146.8±0.7	1.98±0.03	36

Table II. Analysis of Alpha Decay of ^{239}Am to Members of the Favored Rotational Band in ^{235}Np.

FINAL STATES		RELATIVE ABUNDANCE UNCORRECTED FOR ENERGY†			ENERGY CORRECTION FACTOR $P_o(Z,E)$‡	SUMMATION NORMALIZED	EXPERIMENTAL ABUNDANCE
SPIN (K = 5/2)	RELATIVE ENERGY (keV)	L = 0	L = 2	L = 4	(%)	(%)	(%)
5/2	0	1.0	.238	1.08×10^{-4}	52.8	84.7	83.7
7/2	42.6		.317	7.84×10^{-4}	31.4	12.94	13.75
9/2	97.8		.111	1.71×10^{-3}	15.8	2.31	1.98

Hindrance factors for L waves deduced from alpha decay of ^{238}Pu and ^{240}Cm, (Ref. 5).

L = 0	1
L = 2	1.5
L = 4	221

†This is equal to the square of the Clebsch-Gordan coefficient for the transition, (Ref. 23), divided by the hindrance factor.

‡Calculated according to Hyde, (Ref. 20), p. 245.

Table III. Intensities of Gamma Rays in ^{235}Pu per 100 K X-rays.

ENERGY (keV)	SINGLES	COINCIDENCE
49.0±0.1	2.67±0.07	1.74±0.27
34.1±0.1	0.33±0.05	*0.20±0.05

*Estimate using data from a separate coincidence run.

Table IV. Lifetime of keV State in ^{235}Np Using Different Methods of Analysis.

METHOD	HALF-LIFE (nsec)
Newton (12)	6.8±0.3
Laplace Transforms (13)	7.3±0.3
Centroid Shift (11,14) (First Moment)	6.3±0.3
Higher Moments (11,15)	7.2±0.4
Least Square Fit (11)	6.9±1.1
Weighted Average	6.9±0.3

FIGURE CAPTIONS

Fig. 1. ^{239}Am alpha spectrum--computer smoothed.

Fig. 2. Least squares analysis of lifetime measurement on 49 keV state in ^{235}Np.

Fig. 3. Comparison of energy levels in ^{235}Np, ^{237}Np, and ^{239}Np.

Fig. 4. Proposed decay scheme for ^{235}Np.

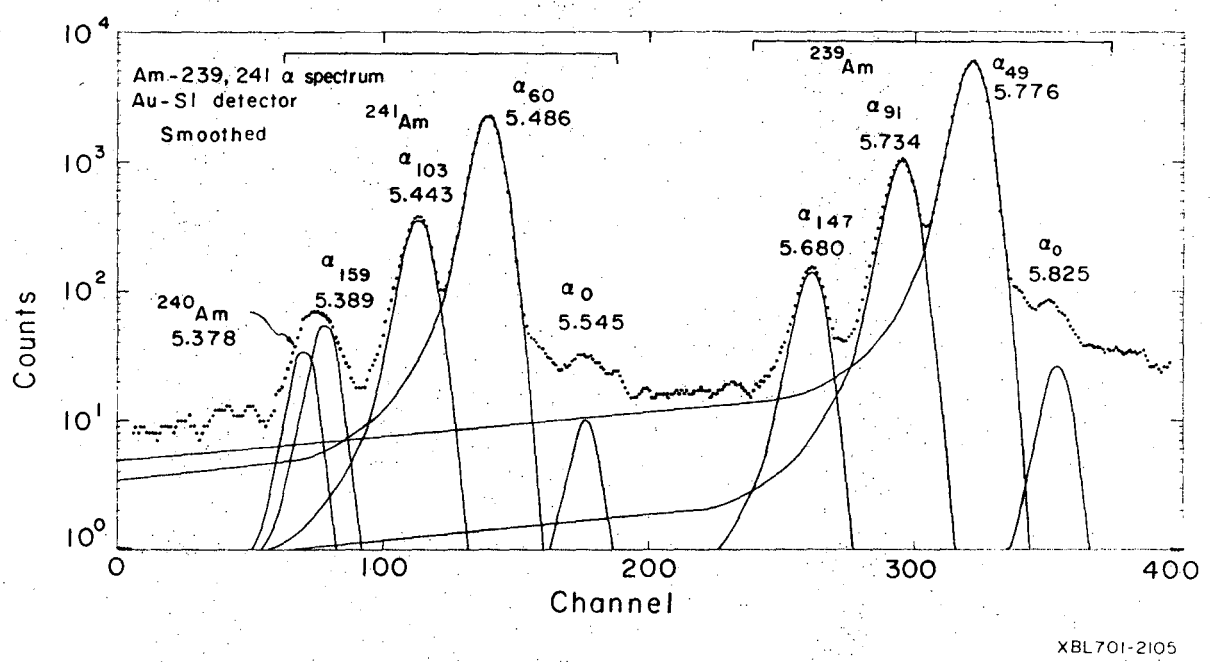
Am-239, 241 α spectrum
Au-Si detector
Smoothed
241Am
239Am
240Am
5.378
α159
5.389
α103
5.443
α60
5.486
α0
5.545
α147
5.680
α91
5.734
α49
5.776
α0
5.825
Counts
Channel
0
100
200
300
400
10^0
10^1
10^2
10^3
10^4
XBL701-2105

Fig. 1

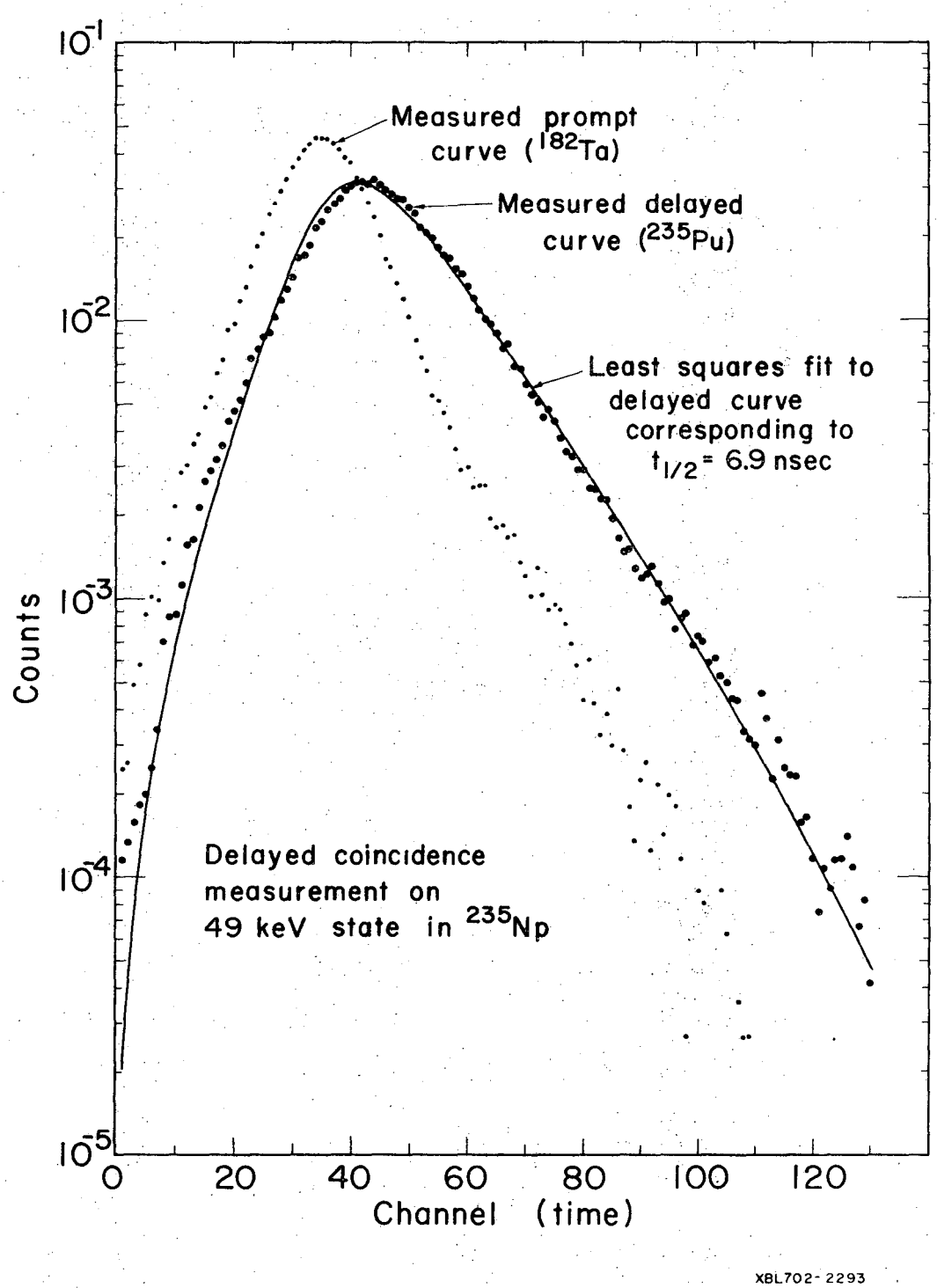
Measured prompt curve (^{182}Ta)
Measured delayed curve (^{235}Pu)
Least squares fit to delayed curve corresponding to $t_{1/2}$ = 6.9 nsec
Delayed coincidence measurement on 49 keV state in ^{235}Np
Counts
Channel (time)
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-5}
0
20
40
60
80
100
120
XBL702-2293

Fig. 2

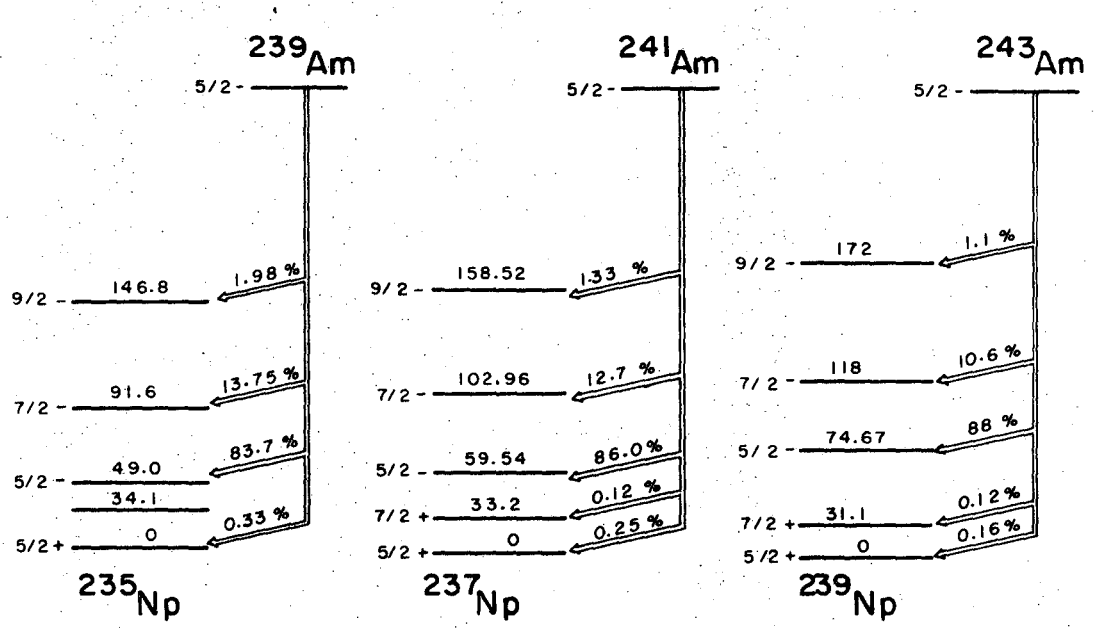

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Fig. 3

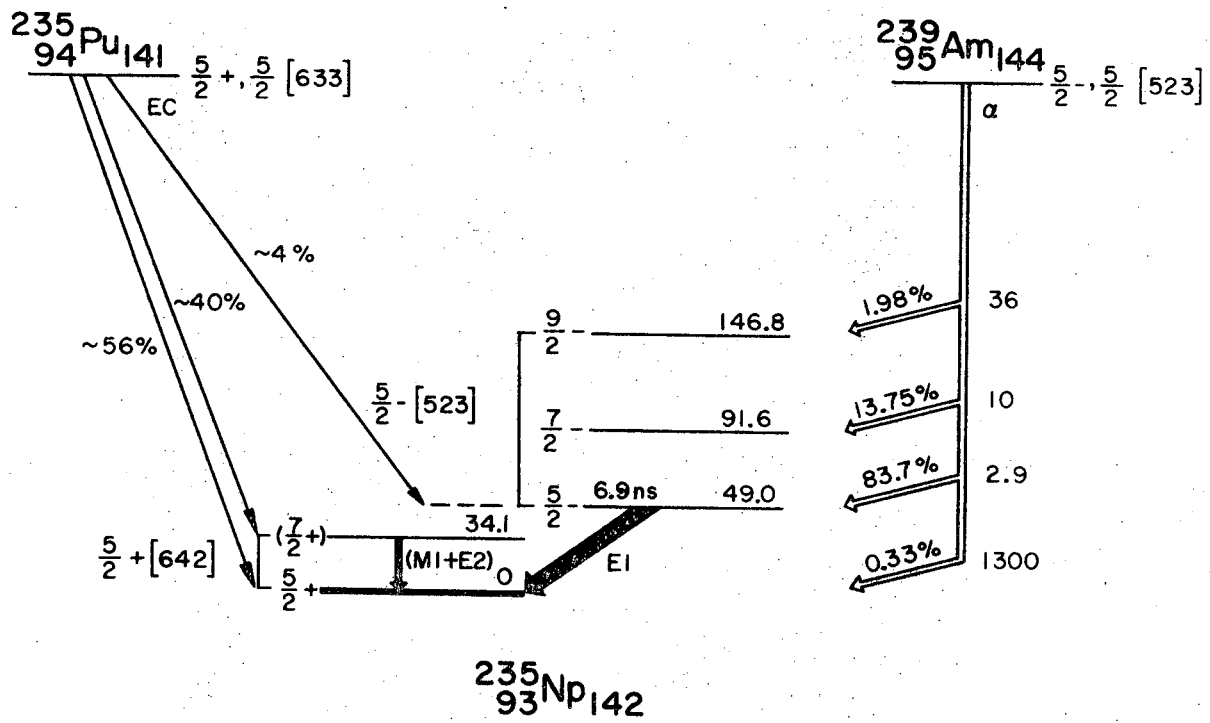
235 94 Pu 141
5/2 +, 5/2 [633]
EC
~4 %
~40%
~56%
5/2 - [523]
5/2 + [642]
(7/2 +)
5/2 +
34.1
(M1+E2)
0
9/2 -
146.8
7/2 -
91.6
5/2 -
6.9ns
49.0
E1
235 93 Np 142
239 95 Am 144
5/2 -, 5/2 [523]
α
1.98%
36
13.75%
10
83.7%
2.9
0.33%
1300
XBL701-2127

Fig. 4

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