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BETA EMMITTER Np^{238}

II. SCINTILLATION SPECTROSCOPY AND COINCIDENCE STUDIES

John O. Rasmussen, Frank S. Stephens, Jr., Donald Strominger, and Björn Aström

February 17, 1955

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ABSTRACT

The relative intensities of electromagnetic radiation from Np^{238}, determined by scintillation spectroscopy, set an upper limit for K capture of K/beta <1 percent.

On the basis of extensive scintillation counter coincidence measurements together with the beta spectroscopic results of the preceding article two alternate decay schemes, differing only in minor detail, are proposed for Np^{238}. The levels of Pu^{238} involved are three close-lying ground rotational band members and a cluster of three levels near 1 Mev, two of which appear to belong to the same rotational band. Conversion coefficient determinations permit multipolarity assignment for a number of the gamma transitions and consequent spin and parity assignments for a number of excited states. Selection rules and other intensity rules involving the Bohr-Mottelson K-quantum numbers are tested, providing K assignments for levels. There is a high degree of K-purity of those states where tests were possible. The question of possible "vibrational" character of the band near 1 Mev is discussed speculatively.

Much of this work was performed under the auspices of the U.S. Atomic Energy Commission.

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INTRODUCTION

Supplementing the beta spectroscopic studies of Np^{238}, as reported in the preceding paper[1] (referred to here as paper I), we have carried out scintillation spectroscopic studies of the electromagnetic radiation and have also performed various coincidence studies with scintillation counting equipment. The coincidence results are of key importance in the establishment of a probable decay scheme.

SCINTILLATION SPECTROSCOPY AND THE QUESTION OF K-CAPTURE

The Np^{238} sample was prepared by thermal neutron bombardment of Np^{237} in the homogeneous reactor at the University of California Radiation Laboratory, Livermore, California. The integrated neutron flux during the irradiation was sufficiently small that no significant amount of Np^{239} was formed. Chemical purification involving lanthanum fluoride precipitation followed by two TTA (thenoyltrifluoroacetone) solvent extraction

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Much of this work was performed under the auspices of the U.S. Atomic Energy Commission.

1. Rasmussen, Slätis, and Passell, Phys. Rev., preceding article.

steps similar to the procedures described by Magnusson, Thompson, and Seaborg[2] was made to purify the neptunium fraction from fission products and other impurities.

The 50-channel pulse-height[3] analyzer was used to study the gamma spectrum of Np^{238} from a sodium iodide scintillation crystal. This spectrum shows an unresolved photopeak near 1 Mev made up of 4 high-energy gamma rays,[1] a photopeak near 100 kev made up of 102-kev gamma and K x-rays, and a low-energy x-ray peak of about 20 kev. Scintillation spectroscopy in Stockholm showed the same peaks and no others.

The apparent resolution (ratio of energy width at half-height to photopeak energy expressed in percent) of this 1-Mev photopeak was 10.6 percent compared with 7.3 percent for the 0.84-Mev photopeak of Mn^{54} and 7.5 percent for the 1.06-Mev photopeak of Bi^{207}. This result clearly shows the composite nature of the 1-Mev peak in Np^{238}.

The 1 Mev photopeak decayed with a half-life of 2.04 days, in excellent agreement with the 2.10-day half-life previously reported by Freedman et al.[4]

The decay of the peak in the 100-kev region was complex. This complexity can be explained by the presence of a considerable quantity of

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2. Magnusson, Thompson, and Seaborg, Phys. Rev. 78, 363 (1950).
3. Ghiorso and Larsh, University of California Radiation Laboratory Report UCRL-2647, 1954 (unpublished).
4. Freedman, Jaffey, and Wagner, Phys. Rev. 79, 410 (1950).

Np^{237} and the growth of its alpha decay daughter, Pa^{233}, into the sample. The decay of the 100-kev peak was followed for 4 months, at the end of which time it was possible to calculate the component contributions due to Np^{238}, Np^{237}, and Pa^{233}.

Corrections were then made for the sodium iodide crystal efficiencies for the 1-Mev and the 100-kev photopeaks.[5] As a result, it was determined that the gamma and K x-radiation of the 100-kev region had a total intensity of 3.8 percent of the total number of unconverted gamma transitions in the 1-Mev region. Only 55 percent of the beta transitions populate the energy levels near 1 Mev in Pu^{238}.[1] Therefore, the observed intensity of the approximately 100-kev photons in the decay of Np^{238} is 2.1 percent of the total beta disintegrations. From beta spectroscopic results we can account for 0.5 percent as K x-rays from conversion of energetic gamma rays and 0.5 percent as 102-kev gamma rays (using a conversion coefficient[6] of 5). There is an excess of 1.1 percent photons not accounted for, and this figure constitutes an upper limit for possible K capture. It seems highly unlikely that the excess is in fact attributable to K capture, since the latest estimate of Glass et al.[7] of the available energy for electron capture (based on

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5. M. I. Kalkstein and J. M. Hollander, University of California Radiation Laboratory Report UCRL-2764 (1954).
6. Asaro, Perlman, and Thompson, Phys. Rev. 92, 694 (1953).
7. Glass, Thompson, and Seaborg, Journal of Inorganic and Nuclear Chemistry 1, 1 (1955).

closed decay cycle energy balances) is 0.10 Mev, insufficient for K capture to occur at all. We attribute the excess 100-kev photons to fluorescence radiation from the Np^{237} mass in the sample.

Jaffe[8] has made a study of the L x-rays arising from the Np^{238} decay, using a Cauchois-type bent-crystal spectrometer of high resolution. His failure to find L x-rays of uranium allows him to set an upper limit of 4 percent for L_I capture and 2 percent for L_{II} capture relative to the beta transitions.

COINCIDENCE MEASUREMENTS

It is well to bear in mind what is known concerning the lowest-lying levels, as determined by studies[6] of the alpha emitter Cm^{242}. There are excited states at 44 and 146 kev with spin and parity assignments of 2+ and 4+, respectively. There are cascade E2 gamma transitions interconnecting these states. From the theoretical conversion coefficients of Gellman et al.,[9] the experimental conversion coefficients of Asaro et al.,[6] and the relative L:MNO conversion results discussed in paper I we estimate that the 44-kev transition is almost totally (99.9 percent) internally converted (77 percent by L_{II} and L_{III} conversion), and the 102-kev transition is approximately 83 percent converted (67 percent of the conversion electrons being from L_{II} and L_{III} orbitals). Thus, for the energetic gamma rays, those going to the ground state will be in coincidence with neither L x-rays nor 102-kev gamma rays. Of those going to the 44-kev state,

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8. H. Jaffe, Ph.D. Thesis. University of California Radiation Laboratory, Report UCRL-2537, 1954 (unpublished).
9. Gellman, Griffith, and Stanley, Phys. Rev. 85, 944 (1952).

about 38 percent will be in coincidence with L x-rays (assuming a fluorescence yield of 50 percent). Of those going to the 146-kev state, approximately 17 percent will be in coincidence with 102-kev gamma radiation, about 45 percent with one L x-ray, and about 11 percent with two L x-rays.

Most of the coincidence measurements were carried out with apparatus employing a sodium iodide (thallium activated) crystal and Dumont 6292 photomultiplier tube in each coincidence channel. A pulse in one channel of a height selectable by a single-channel differential discriminator generates a gate pulse which gates a 50-channel pulse-height analyzer[3] to analyze a simultaneous pulse in the other coincidence channel. The effective coincidence resolving time, $2\tau_o$, was about 7 microseconds for these experiments. The sample was placed directly between the two detectors, spaced such that the 1-inch cylindrical detector crystals each subtended about 30 percent of 4π solid angle. The detectors were shielded by lead.

The energetic gamma spectrum in coincidence with electromagnetic radiation approximately 100 kev in energy (including K x-rays and 102-kev nuclear gamma rays) was examined. In order to prevent beta particles from registering as gate counts the 100-kev radiation was observed through 691 mg/cm^2 of aluminum. In addition 4 gm/cm^2 of lead was used on the signal side of the coincidence unit, since this amount of absorber is not sufficient to cause serious attenuation of the hard gamma rays.

The curve of Fig. 1a shows the coincident gamma energy spectrum observed. This is to be compared with the ordinary total gamma ("singles") spectrum (solid line lower curve) taken at the same amplification. The shift of the peaks between the two curves is to be noted, the peak in

coincidence with about 100-kev radiation lying at the low-energy side of the composite "singles" peak. We know from the beta spectroscopic results in paper I that the gamma "singles" peak consists of at least four different gamma rays of energies 1029, 986, 942, and 927 kev. From these coincidence results we conclude that γ_{942}, γ_{927}, or both, proceed to the 146-kev level. This result was reported earlier.[10] From the beta spectroscopic results we know that only the four high-energy gamma rays have enough energy for K conversion. Thus, there can be no interference in the above coincidence measurement from K x-ray-gamma coincidences.

The peak shift result for the radiation coincident with about 100-kev radiation was independently determined using another coincidence-pulse analysis apparatus with a resolving time $2\tau_o$ of about 6×10^{-8} second. With the shorter resolving time a more active sample could be used. The detection crystals were further from the sample and making a 90° angle with it. Two inches of lead shielding was placed between the crystals to minimize spurious coincidences arising from Compton scattering of gamma rays from one crystal to another. The counters were unshielded except for the one lead brick between them. Sufficient beryllium absorbers to stop completely all beta rays were used before each detector. Thus, we feel that these results, though concerning a rare coincidence process, are real and not spurious.

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10. Rasmussen, Passell, and Stephens, University of California Radiation Laboratory Unclassified Report UCRL-2585, 1954 (unpublished).

The energetic gamma spectrum in coincidence with L x-rays (about 18 kev) was studied with the first mentioned coincidence equipment. In this case it was not possible to use aluminum to absorb the beta particles, as it would attenuate the L x-rays by too large a factor. Therefore 797 mg/cm^2 of beryllium was used on the gate side of the coincidence unit. The hard gamma rays were observed through 2.9 gm/cm^2 of lead.

The dashed curve of Fig. 1b is the resulting coincidence spectrum and is plotted together with the ordinary "singles" gamma spectrum to show most clearly their differences. The curves are arbitrarily normalized to be equal at channel 19. One notes that the high energy side of the broad "singles" peak is strongly depressed in the coincidence spectrum, a clear proof that the maximum energy gamma ray, γ_{1029}, proceeds to ground. (We shall later propose in the decay scheme section that γ_{1029} is actually two gamma rays, within 2 kev of the same energy, the more intense of which goes to ground and the other to the 44-kev state. The curves of Fig. 1b are consistent with such an interpretation.) One also notes that the lower energy part of the "singles" peak is slightly enhanced in the L x-ray coincidence spectrum, a further confirmation of the earlier conclusion that γ_{927}, γ_{942}, or both proceed to the 146-kev state.

A determination was also made of the absolute L-gamma coincidence rate, which measurement is of great aid in establishing intensities of various radiations in the decay scheme. That is, the single channel analyzer was set to accept pulses from the photopeak of the energetic gamma rays, and the total number of gates generated by the single channel analyzer in a given counting time was recorded along with the integrated L x-ray coincidence peak recorded on the 50-channel analyzer. A similar

measurement on alpha particle-L x-ray coincidences for Cm^{242} was made with the same geometries and absorption path (aluminum backing) for L x-rays. By comparing the results of the two measurements (knowing from alpha spectroscopy[6] that 26.3 percent of the alpha particles populate the 44-kev state and 73.7 percent, the ground state), it was possible to calculate the fraction of energetic gamma rays populating the ground state, the calculation being independent of such uncertain quantities as L fluorescence yield, L x-ray absorption loss, and detector geometry. For such a calculation we make use of the result from the beta spectroscopic work reported in paper I that the 146-kev state is populated in 3 percent of the beta disintegrations and that energetic gamma transitions occur in 55 percent of the disintegrations. Also, the conversion coefficients for the 102-kev gamma ray are used.

The result of the determination may be stated in the following two ways:

If there were no energetic gamma rays going to the 146-kev state, the result would indicate 37 percent (based on total beta disintegration) of the gamma rays going to the 44-kev state and 18 percent going to ground.

If we assume the 146-kev state is populated entirely by gamma and not by beta transitions, the proportion of high energy gamma rays going to the various states (percentage of total beta disintegrations) must be 3 percent to the 146-kev state, 32 percent to the 44-kev state, and 20 percent to ground.

A coincidence measurement similar to the preceding one was made between electrons and L x-rays, with the substitution of an anthracene crystal detector for the electrons. We wished to determine the fraction

of hard beta disintegrations (E_{max} = 1.25 Mev) which went to the various low-lying states. Electron energies including sections of the hard beta spectrum but avoiding conversion lines and the soft beta spectra (E_{max} = 0.27 Mev) were selected by the single-channel analyzer serving as the coincidence gate generator. The ratio of L x-ray coincidence counts to gates was measured and again compared with the alpha-L x-ray results from a Cm^{242} standard in the same position. The result of such a measurement was reported[10] previously as indicating (assuming no hard beta decay to the 146-kev state) that 72 percent of the hard beta transitions go to the 44-kev state, and 28 percent to ground. Later results showed the first report to be in error. The early measurement was made with only one sample, of relatively high activity, and there is apparently a loss in coincidence efficiency at higher counting rates in this apparatus. The later results showed a counting value of 100 percent of hard beta particles to the 44-kev state when sufficiently small samples were used. Beta particle energies of 410 to 610 and 1020 to 1200 kev were used as gates and both intervals showed the above limit. (It should be mentioned regarding the loss effect that the L-gamma results of the preceding paragraph were checked with sources of strengths differing by a factor of four and found essentially constant.)

Engelkemeier was reported by Freedman, Jaffey, and Wagner[4] to have found coincidences between hard electrons and electromagnetic radiation of about 100-kev energy. The determination is an important one, as it bears on the question of whether a hard beta group populates the 146-kev state. Our gamma-gamma coincidence work showed that the 146-kev state is populated by energetic gamma rays, but to what fraction of its 3 percent

total population was not determined. It is clear that there will be coincidences between K x-rays and energetic K conversion lines (0.5 percent of total disintegrations), but by examining the energy spectrum of electrons in coincidence with 100-kev radiation it was thought that it might be possible to find a hard beta continuum under the conversion lines. It is clear that the coincidence rate with a hard beta continuum will be about equal to the conversion line coincidence rate if all the 3 percent population of the 146-kev level were due to a hard beta group.

Figure 2 shows the results of such a measurement of the electron spectrum in coincidence with 100-kev radiation. The data are probably consistent with there being no hard beta continuum at all, but drawing in a curve for the maximum amount of hard beta continuum that could be present, one can set an upper limit that coincidences with a continuum must be less than 80 percent of coincidences with conversion electrons. The limit only tells us that the 146-kev level cannot be completely populated by the hard beta group. We shall arbitrarily assume for the later decay scheme proposals that the 146-kev level is populated entirely by energetic gamma rays and not by beta particles.

DECAY SCHEME PROPOSALS

In attempting to construct a decay scheme we start first with the ground rotational band known[6] from alpha decay fine structure studies in Cm^{242}. That is, there are levels in Pu^{238} at 0, 44.0, and 146.2 kev with spin and parity 0+, 2+, 4+, respectively. Connecting these levels are cascading E2 transitions, highly internally converted.

There are four high energy gamma transitions of 927, 942, 986, and 1029 kev to account for. The L-gamma coincidence result that γ_{1029} goes to ground means there must be a level at 1029 kev. The 986-kev gamma transition is then accounted for as the transition between the 1029-kev level and the 44-kev level. The hard gamma-102-kev gamma coincidence result was that γ_{927}, γ_{942}, or both are in coincidence with 102-kev radiation, and the hard gamma-L x-ray coincidence studies (hard gamma peak shape comparisons) tend to support the conclusion that neither γ_{927} nor γ_{942} go to ground. Thus, regarding additional levels, we are left with three choices: 1) levels at 1073 and 1087 kev, 2) levels at 1073 and 986 kev, or 3) levels at 1087 and 971 kev. It is to be noted with the first two choices that there are two separate places in the decay scheme where a gamma transition of 1029 kev could occur, namely, from the 1029-kev level to ground and from the 1073-kev level to the first excited state. In order to reconcile the numerical L-gamma coincidence result that the hard gamma intensity to the ground state is 20 percent of total beta disintegrations, we can postulate that the 1029-kev gamma is indeed complex with energy difference of the two components too small for resolution even on the double-focusing beta spectrometers. Therefore, we prefer alternatives (1) and (2) for our further decay scheme discussions.

The attempt to assign intensities to all the transitions in the decay scheme is handicapped by incomplete information regarding intensities of the weaker hard gamma and soft beta transitions. Nevertheless, by using

the absolute L-gamma coincidence result of 20 percent hard gamma to ground, the uranium photoconversion electron result of paper I that $\Gamma_{1029}:\Gamma_{986} = 1:0.83$, and other intensities from the beta spectroscopic work (102-kev transition = 3 percent; hard beta:total soft beta = 45:55) and assuming intensities for γ_{927} and γ_{942} in the ratio of their K conversion lines, 1:2, we can calculate intensity figures for all transitions with decay scheme (1). It is obvious that the intensities of the weaker transitions as determined in this manner may be very uncertain where they are based on differences of relatively large numbers. Figure 3 shows the resulting decay scheme, where the uncertain intensities are given in parentheses. Below the intensities of the various beta groups are given their log ft values, underlined.

Using the same intensity data but with the level scheme discussed as alternative (2) above, one obtains the decay scheme of Fig. 4. With the scheme of Fig. 4 it is possible that γ_{986} might also be complex, as there are two places in the scheme where it might occur. The purely arbitrary assumption of zero intensity for the transition from the 986-kev level to ground was made for Fig. 4.

While either decay scheme accounts for most of the data, it is appropriate to summarize the remaining inconsistencies.

First, according to the decay schemes the 44-kev transitions should occur in 80 percent of the total beta disintegrations, the percentage having been fixed by the absolute L x-ray-hard gamma coincidence rate measurement. In considerable disagreement is the figure of 65 percent from the beta spectroscopic measurements of Slätis, Rasmussen, and Atterling.[11]

11. Slätis, Rasmussen, and Atterling, Phys. Rev. 93, 646 (1954).

Even the figure of 75 percent from Freedman, Jaffey, and Wagner[4] is on the low side. Possible sources of error in the electron spectroscopic intensities could result as a consequence of the low energy of conversion lines, with uncertainty in counter window transmission corrections and resolution of peaks broadened by finite sample thickness. The ratios of intensities of conversion lines (L_{II}, L_{III}, M, N) determined by Slätis et al.[11] agree better than those of Freedman et al.[4] with independent determinations of Passell.[12] All these measurements are compared in Table I of the preceding paper I. The true intensity of gamma rays to ground is probably intermediate between the coincidence and the spectroscopic determinations, but in view of the disagreement between the different spectroscopic determinations we have chosen to use the coincidence measurement as the basis for the decay scheme intensities.

Second, it is to be noted that the absolute relative amounts of hard gamma and approximately 100-kev radiation as determined by scintillation pulse analysis do indicate an excess of approximately 100-kev radiation beyond that given by the proposed decay schemes. It is not reasonable on energetic grounds, as discussed earlier, to attribute this excess radiation to K capture. A related discrepancy involves an apparent excess over expectation of soft beta coincidences with approximately 100-kev radiation, as measured roughly by integration of the curve of Fig. 2. The measurement is not very quantitative, since scattering and absorption effects on the electrons in this coincidence experiment introduce uncertainties that would be hard to estimate.

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12. T. O. Passell, Ph.D. Thesis, University of California Radiation Laboratory Report UCRL-2528, 1954 (unpublished).

Finally, regarding further work which might be profitable, we would suggest that beta spectroscopic work at even higher resolution might be fruitful, especially in the region of the high energy conversion lines. Coincidence experiments with two beta spectrometers would be desirable. Experiments to determine the relative intensities of the weaker hard gamma rays are important. Such might lie within the range of Cauchois-type bent crystal spectrometers, or one might study by beta spectroscopy the K photoelectrons from radiators of considerably lower atomic number than uranium. A uranium radiator was used in the experiments reported in paper I, and the K photopeaks of the weaker gamma rays lie obscured by Compton electrons of the intense gamma rays.

HARD GAMMA CONVERSION COEFFICIENTS

Using the gamma intensities from the decay schemes and the K electron line intensities from paper I, the K conversion coefficients of the hard gamma rays have been calculated. Table 1 summarizes the data and results.

It is felt that a multipolarity assignment on the basis of K conversion coefficients for the two most intense hard gamma rays is justified. Despite the probable unresolved doublet nature of γ_{1029} and the consequent average meaning of its conversion coefficient, the major branch of the doublet is probably of E2 character, necessarily pure E2 since the final state has spin zero. Thus, the level at 1030 kev must have spin 2 and even parity, as the ground state is presumably 0+. The fact that the experimental conversion coefficient is smaller than the theoretical unscreened value may in part be due to experimental uncertainty and in part due to neglect of screening in the theoretical calculations. With regard to the latter point, some of the

Table 1

Hard Gamma Conversion Coefficients

Gamma energy	Gamma intensity (%) Scheme 1	Gamma intensity (%) Scheme 2	K line intensity (%)(from Berkeley double-focusing spectrometer)	Experimental K conversion coefficient Scheme 1	Experimental K conversion coefficient Scheme 2	Theoretical K conversion coefficients (interpolated from Rose et al.[13]) E1	E2	M1	M2	Experimental K:L ratio (from Berkeley double-focusing spectrometer)	Multipolarity assignment
Σ1029	28	24	0.20	0.0071	0.0083	0.0033	0.0102	0.075	0.105	~5	E2
986	24	21	0.20	0.0083	0.0095	0.0035	0.0108	0.083	0.117	~3.3	E2
942	(2)	(7)	0.07	(0.035)	(0.010)	0.0038	0.0119	0.094	0.132	--	--
927	(1)	(3)	0.04	(0.04)	(0.013)	0.0039	0.0122	0.098	0.136	--	--

13. Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. 83, 79 (1951).

recent screened K conversion coefficients calculated by Rose and others[14] seem roughly 10 percent lower than the unscreened for E2 transitions of comparable energy in large Z elements.

Within experimental uncertainty the conversion coefficient of γ_{986} indicates E2, although an admixture of a few percent M1 character could not be excluded by our data.

The uncertainties associated with the gamma radiation intensities of the weaker hard gamma rays make definite multipolarity assignments out of the question, but their orders of magnitude indicate E2 or mixed E2-M1 character as most likely. (It is apparent that with all the gamma rays the conversion coefficients might be explained by E1-M2 mixtures, but this explanation is not given serious consideration in view of the general rarity of that particular admixture, and the K/L ratios give experimental evidence against the E1-M2 interpretation.)

It is of some interest to compare the K/L conversion ratios with the earlier correlation of Goldhaber and Sunyar.[15] For gamma rays about 1 Mev and Z = 94 their plots against Z^2/E would give a K/L ratio of about 6 for E2 and about 8 for M1 or M2. The lower experimental K/L ratios in the case of our E2 gamma rays of 1029 and 986 kev are further examples of systematic deviations to the low side for high Z elements. Goldhaber and Sunyar[15] called attention to some such deviations, and the work of

14. M. E. Rose, privately circulated tables (1954).

15. M. Goldhaber and A. W. Sunyar, Phys. Rev. 83, 906 (1951).

Alburger and Pryce[16] on Bi^{206} decay provides a good example of this sort of deviation. Their E2 gamma transitions of 880.5 and 803.3 kev have $K/(L_I + L_{II})$ ratios of about 4.7. Our K/L ratios from Table 1 are consistent with the E2 assignment in view of the considerable uncertainty in our L line intensities. From the L subshell conversion calculations of Gellman et al.[9] inclusion of the L_{III} conversion in K/L ratios would have virtually no effect for M1 transitions and less than 30 percent effect for E2. Thus, we are justified in comparing our K/L ratios with $K/(L_I + L_{II})$ ratios.

CONSIDERATIONS INVOLVING THE K QUANTUM NUMBERS

Two interesting observations may be made with regard to the gamma transitions occuring from the 1030-kev level. First, the rather pure E2 character of γ_{986}, though M1 is allowed, seems unusual according to lifetime estimates from the single particle lifetime formulas. Second, the absence (see paper I) of an E2 transition of 884-kev energy proceeding to the 4+ state at 146.2 kev seems unusual. It is concerning just such observations as these (relative transition rates to various members of a nuclear rotational band) that the deformed nuclear model makes precise predictions, and one might expect the model to be useful around mass number 238 where nuclear rotational band systems are much in evidence. For the present we need consider only the intensity rules arising from the simple approximate separation of the nuclear motion into the relatively

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16. D. E. Alburger and M. H. L. Pryce, Phys. Rev. 95, 1482 (1954). (See Fig. 10.)

slow rotational motion of a spheroidally deformed nucleus and a more rapid internal motion of a deformed intrinsic structure. This separation is described in the forthcoming paper of Alaga, Alder, Bohr, and Mottelson[17] and constitutes a more general case of the strongly deformed nuclear model of Bohr and Mottelson.[18] The rotational separation introduces one quantum number in addition to I, the total angular momentum, and π, the parity. The new quantum number is K, the projection of the total angular momentum along the nuclear symmetry axis, and the K-purity of nuclear states will depend upon how valid is the approximate separation of rotational and intrinsic structural motion. K remains the same for all members of the same rotational band and is equal to the spin of the base level of the rotational band except for an anomalous case when K = 1/2. Thus, K is zero for the members of the ground rotational band as indicated in Figs. 3 and 4.

The relative rates of the electric quadrupole transitions from the 1030-kev level to the states of the ground rotational band give us a means of evaluating K for the 1030-kev level. After dividing out the fifth power energy dependence we may obtain the ratios of reduced transition probabilities. By the theory these ratios should be simply given by vector projection factors, the squares of Clebsch-Gordan coefficients involving the K and I quantum numbers and the multipolarity L of the transition. This relation as given by equation (12) of Alaga et al.[17] is shown below:

$$\frac{B(L, I_i \rightarrow I_f)}{B(L, I_i \rightarrow I_f')} = \frac{\langle I_i L K_i K_f - K_i \mid I_i L I_f K_f \rangle^2}{\langle I_i L K_i K_f - K_i \mid I_i L I_f' K_f \rangle^2}$$

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17. Alaga, Alder, Bohr, and Mottelson, CERN/T/A-A-B-M-1 (August 1954), (to be published in Dan. Mat. Fys. Medd.).

18. A. Bohr and B. R. Mottelson, Dan. Mat. Fys. Medd. 27, (16) (1953).

where the Clebsch-Gordan coefficients are in the notation of Condon and Shortley.[19] Clebsch-Gordan coefficients have been tabulated numerically by Simon.[20] Table 2 presents the experimental and theoretical (K_i = 0, 1 or 2) relative reduced transition probabilities for the gamma rays from the 1030-kev level.

The assignment K = 2 is clearly indicated for the 1030-kev level. With this assignment the absence of M1 character in γ_{986} finds a natural explanation. The Clebsch-Gordan coefficient vanishes when ΔK exceeds the multipolarity L; hence, dipole radiation is K-forbidden (see Alaga et al.,[17] equation (6a)).

Table 2

Experimental and Theoretical Relative Reduced E2 Transition Probabilities from the 1030-kev level (2+)

Final state I_f	Experimental $\frac{B(2 \to I_f)}{B(2 \to 0)}$		Theoretical $\frac{B(2 \to I_f)}{B(2 \to 0)}$		
	Scheme 1	Scheme 2	$K_i = 0$	$K_i = 1$	$K_i = 2$
2+ (44 kev)	1.5	1.3	1.43	0.375	1.43
4+ (146 kev)	<0.3	<0.3	2.57	1.14	0.0714

19. Condon and Shortley, Theory of Atomic Spectra, Cambridge University Press, London, 1935.

20. Albert Simon, Oak Ridge Laboratory Report ORNL-1718, special (1954).

The spacing of 44-kev between the 1029- and 1073-kev levels and the gamma radiation from the 1073-kev level suggest to us that the 1073-kev level be assigned (2, 3+) as the first excited member of a rotational band based on the 1029 level. The rotational moment of inertia for this upper band is the same as that of the ground state band within the accuracy of our measurements. The theory predicts (for pure E2 radiation) $B(I_i = 3 \rightarrow I_f = 4)/B(I_i = 3 \rightarrow I_f = 2) = 0.40$. The experimental relative intensities of the gamma rays from the 1073-kev state are too uncertain to make a significant test of the reduced transition probability rules discussed above.

INTERPRETATION OF BETA TRANSITIONS

The log ft values of the various beta groups, as calculated using the nomograms of Moszkowski[21] are given on the decay schemes Figs. 3 and 4 as underlined numbers. They are summarized in Table 3.

Table 3

Beta Group Energies and Log ft Values

Beta Energy (kev)	K, spin and parity of final state	$Log_{10}ft$ Scheme 1	$Log_{10}ft$ Scheme 2
1260	0, 2+	8.5	8.5
275	2, 2+	5.8	5.8
231	(2, 3+)	(6.4)	(6.5)
217	?	(7.1)	--
or			
318	?	--	(7.2)

21. S. A. Moszkowski, Phys. Rev. 82, 35 (1951).

Freedman et al.[4] found an allowed spectral shape for the hard beta particle. This result was confirmed in the beta spectroscopic studies of paper I. Parts of the spectrum are obscured by soft beta groups and by conversion lines, but the assumption of allowed shape gives a satisfactory fit, whereas the assumption of the unique alpha shape ($\Delta I = 2$, yes) does not. Thus, a $\Delta I = 2$, yes, interpretation of the hard beta group seems ruled out.

It is somewhat surprising that the 1260- and 275-kev beta groups, which proceed to levels of the same spin and parity, should exhibit ft values different by a factor of 500. Since the relative gamma transition rates suggested that K is a fairly good quantum number, we might logically look first to the difference in K values of the final states at 44 and 1029 kev for an explanation of the great difference in log ft values.

The simplest interpretation in terms of K selection rules follows from an assignment of K, I, and π of (2, 2+) for Np^{238}. The transitions to ground and 146-kev state are not observed as they are second forbidden ($\Delta I = 2$, no). Though the good selection rules in I and π make the transition to the 44-kev state allowed ($\Delta I = 0$, no), the selection rule[17] in the approximate quantum number K makes the transition second forbidden ($\Delta K = 2$, no), and if K for the initial and final states were an absolutely good quantum number the beta transition would not be seen (log ft $\gtrsim 12$). There are evidently small admixtures of states with K values other than the principal ones in the initial state, final state or both, and for some combinations of these K impurities the change in K will be 0, or 1, permitting the transition to proceed as an allowed transition. The allowed spectrum shape of the hard beta group is a natural consequence of this interpretation.

With the Np^{238} (2, 2+) assignment the beta groups to the 1030- and 1073-kev levels, which are (2, 2+) and (2, 3+), respectively, should be allowed transitions by ΔK as well as ordinary selection rules. Their log ft values are a little high (5.8 and 6.5, respectively), the log ft correlations of Nordheim[22] showing all allowed transitions (21 cases) in the log ft range 4.1 to 5.7. There are, of course, cases where other factors in the nuclear rearrangement accompanying beta decay act to make allowed transitions much slower than ordinary. Such a case is our hard beta group, and de-Shalit and Goldhaber[23] have cited and discussed a number of other cases. Thus, the interpretation of our two main soft beta groups as somewhat slow allowed transitions seems not unreasonable.

The assignment of (2, 2+) for Np^{238} cannot be claimed to be unique. One might make an assignment of (2, 2-) but would then need to attribute the non-observance of transitions to the ground state and second excited state to some unknown hindering factor. The log ft values of the 275- and 231-kev beta groups seem a little low (King and Peaslee[24] find average log ft values of 6.5 for $\Delta I = 0$, yes, and 7.5 for $\Delta I = 1$, yes transitions) for the first forbidden nature they would have in this alternate interpretation. One cannot rule out possible assignments of (3, 3$\pm$) to Np^{238}, although with such there would be some difficulty in explaining why beta transitions strongly favor the first excited state (2+) over the second excited state (4+).

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22. L. W. Nordheim, Revs. Modern Phys. 23, 322 (1951).

23. A. deShalit and M. Goldhaber, Phys. Rev. 92 1211 (1953).

24. R. W. King and D. C. Peaslee, Phys. Rev. 94, 1284 (1954).

An assignment of even parity to Np^{238} violates the shell model rule that the ground states of odd-odd nuclei in this region should have odd parity, the rule following from the fact that mainly odd parity orbitals are available for protons in the 82-126 shell and even parity orbitals for neutrons in the shell above 126. We feel, however, that the shell model parity rule is hardly to be trusted in the region of the heaviest elements where numerous low energy E1 transitions to ground in odd A nuclei indicate the ready availability of orbitals of both parities for odd nucleons.

CONCERNING THE POSSIBLE VIBRATIONAL NATURE OF SOME EXCITED LEVELS OF Pu^{238}

Inasmuch as the intensity rules involving K seem useful in the case of Np^{238} decay, the question is bound to be raised whether any of the high levels in Pu^{238} are "vibrational levels," as predicted by the more detailed unified model of Bohr and Mottelson.[18] The question is a difficult one to answer with the data available at present, and what follows is written in a speculative vein.

The theory predicts for spheroidal nuclei two vibrational bands, each having one quantum of vibrational excitation. The so-called beta vibrational band will have rotational band members (0, 0+), (0, 2+), (0, 4+), ... The gamma vibrational band will have members (2, 2+), (2, 3+), (2, 4+), ... The vibrational quantum energy may be of the order of 1 Mev for the heaviest elements. Thus, the levels in Pu^{238} of 1029 and 1073 kev with (2, 2+) and (2, 3+) assignments, respectively, might possibly be of the first gamma vibrational band, although they can equally well be purely "particle excitation" levels.

The most definitive evidence for the vibrational character of the band would be lifetime measurements of the levels. If lifetimes were found to be substantially shorter than predicted by the individual particle lifetime formulas, this would constitute strong evidence for some vibrational character for the levels. Such measurements by ordinary electronic means seem impossible, as the expected lifetimes are so short. Following are the theoretical calculated mean lifetimes for E2 transitions from the (2, 3+) state at 1073 kev: single proton transition to (0, 2+) state at 44 kev, $\tau_{sp} \sim 5 \times 10^{-12}$ sec.; single photon (vibrational transition) to 44-kev state, $\tau_{vib} \sim 3 \times 10^{-14}$ sec.; rotational transition to (2, 2+) state at 1029 kev (taking internal conversion into account), $\tau_{rotational} \sim 5 \times 10^{-11}$ sec. (Lifetime formulas of Bohr and Mottelson[18] were used.) It seems surprising that despite the great energy difference the 44-kev rotational transition could compete to the extent of about 10 percent with the 1029-kev single particle transition. From our data the presence of such competition could not quite have been determined, and we are unable to obtain any lifetime evidence in this manner.

Alaga et al.[17] in their discussion of the beta decay scheme of Ta^{182} cite as possible evidence for the gamma vibrational character of a band at 1222 kev the fact that the rotational moment of inertia for the band is slightly smaller (~10 percent) than that for the ground rota ional band, whereas a particle excitation level generally has a greater rotational moment of inertia. In the case of Pu^{238} the rotational moment of inertia of the upper band is about the same as that of the ground band and constitutes no clear evidence for or against the vibrational assignment.

The log ft values for beta decay to the 1029-kev band in Pu^{238} give further evidence bearing on the question of vibrational character. If the

band were purely a gamma vibrational band with the individual nucleons remaining in paired configurations, a selection rule in the Ω quantum number* makes the principal soft beta transitions from (2, 2+) Np^{238} second forbidden ($\Delta\Omega = 2$, no). The observed ft values indicate the beta transitions are only about a factor of 10 slower than average for allowed transitions. Clearly the Pu^{238} band in question cannot be of very pure vibrational character, if, indeed, it has vibrational character at all. One might speculate that for some levels the separation of the intrinsic structure of the deformed nucleus into vibrational and individual particle motion is just partially valid and could lead to states with partial vibrational character giving enhanced E2 transition rates but with Ω not a very good quantum number.

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*Ω is the component of total individual particle angular momentum along the nuclear symmetry axis.

FIGURE CAPTIONS

Fig. 1. Scintillation spectra (sodium iodide) of the composite 1-Mev gamma peak. Curve la ___ is the spectrum in coincidence with ≈100-kev electromagnetic radiation. Curve lb, dashed line, triangles, _____ is the spectrum in coincidence with L x-radiation. Curve lb, solid line, circles, ___ is the total gamma spectrum. The curves lb are normalized at channel 19 (the position of the 986-kev gamma ray) to demonstrate best the differences between the curves. The marks on the baseline of la indicate the expected peak positions of the various component gamma rays, based on energy calibration with Bi^{207} and Mn^{54} standards.

Fig. 2. Scintillation spectrum (anthracene) of electrons in coincidence with ~100-kev electromagnetic radiation.

Fig. 3. Possible decay scheme (first of two alternative schemes) for Np^{238}. Radiation intensities are given as percentage of total beta disintegrations, and intensities enclosed in parentheses are indirectly determined and subject to considerable uncertainty. Below the beta group intensity figures are underlined numbers, the log ft values.

Fig. 4. Possible decay scheme (second of two alternative schemes) for Np^{238}. Radiation intensities are given as percentage of total beta disintegrations, and intensities enclosed in parentheses are indirectly determined and subject to considerable uncertainty. Below the beta group intensity figures are underlined numbers, the log ft values.

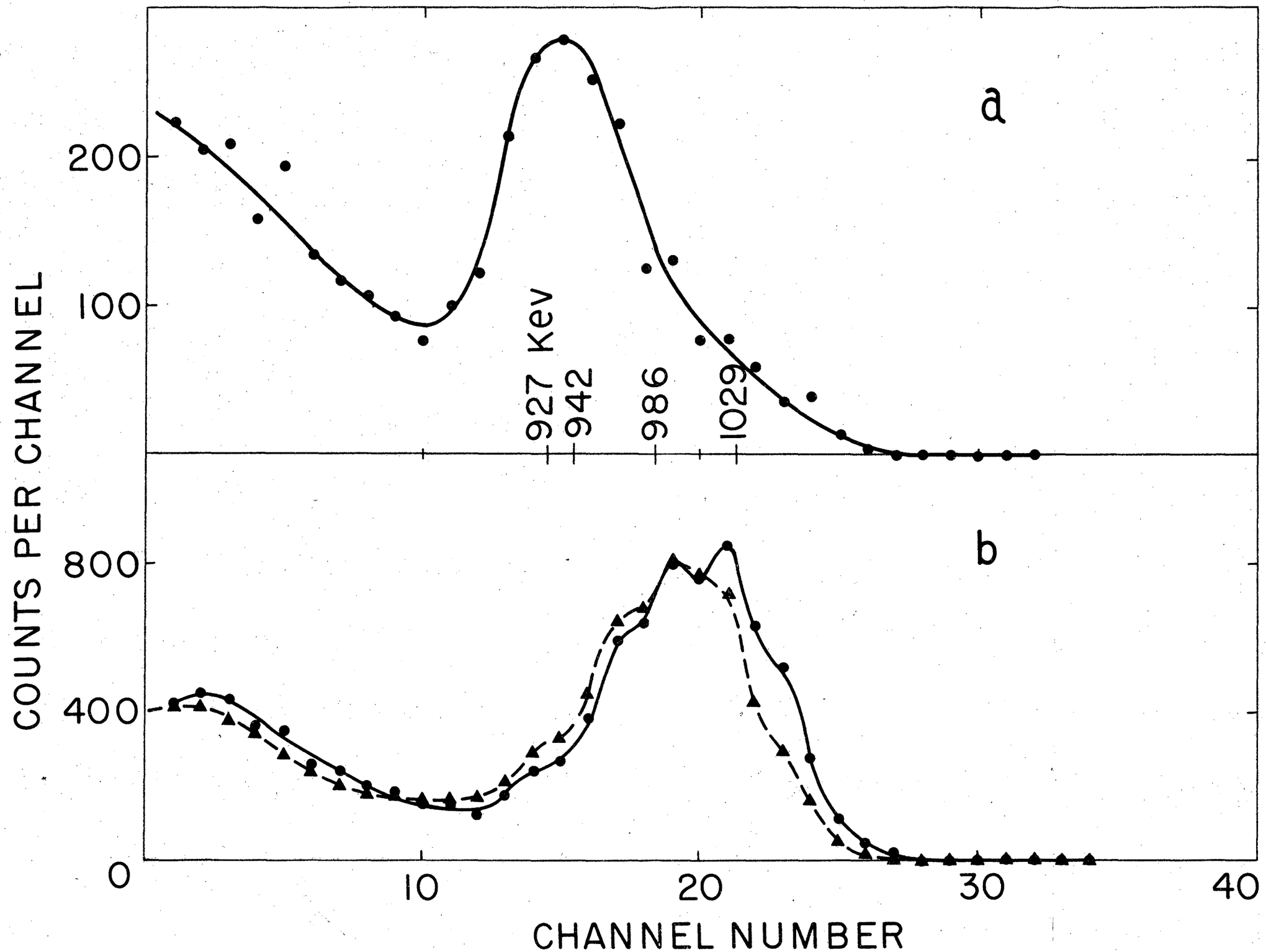

a
b
200
100
800
400
0
10
20
30
40
927 Kev
942
986
1029
COUNTS PER CHANNEL
CHANNEL NUMBER

Fig. 1 23038-2

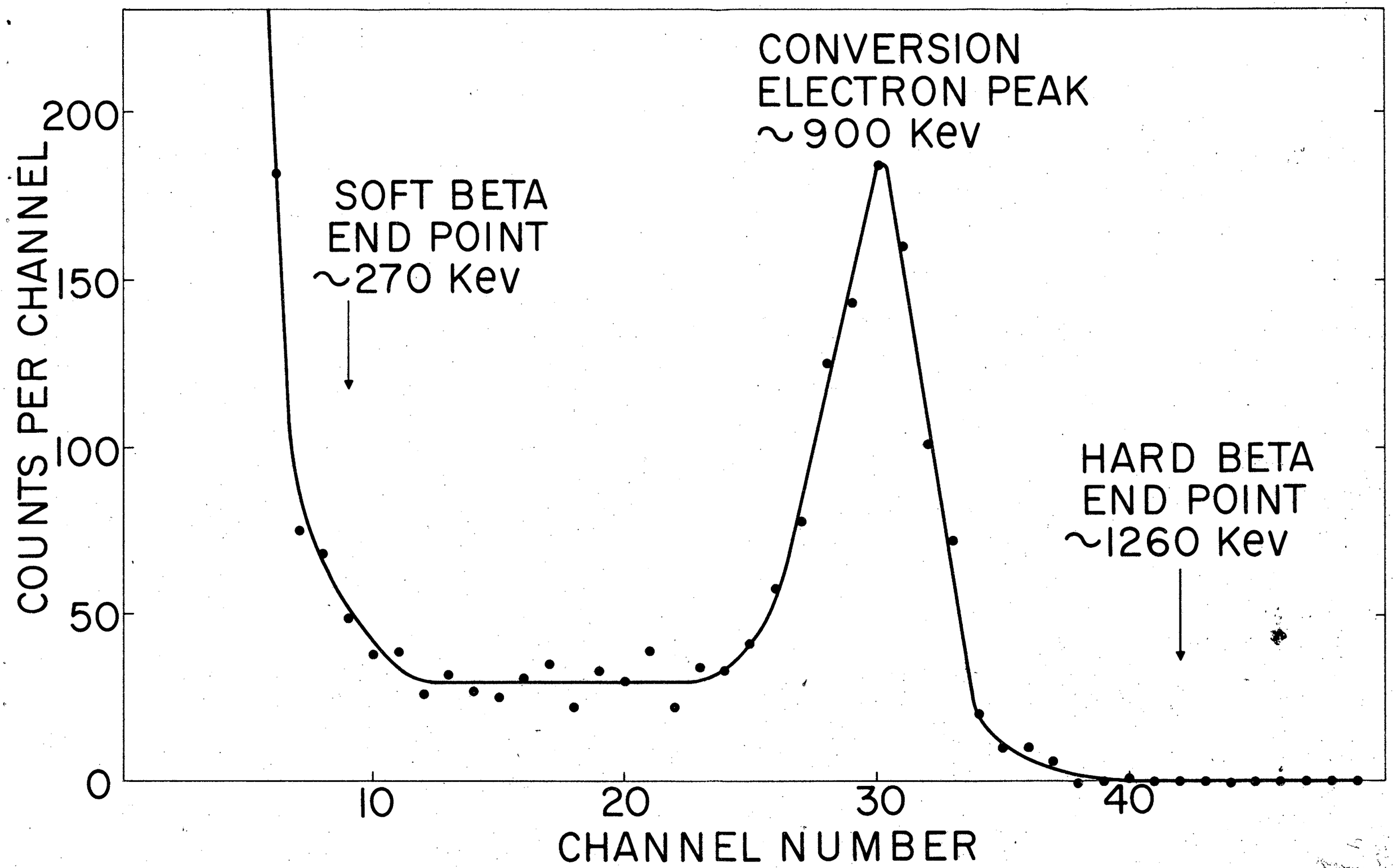
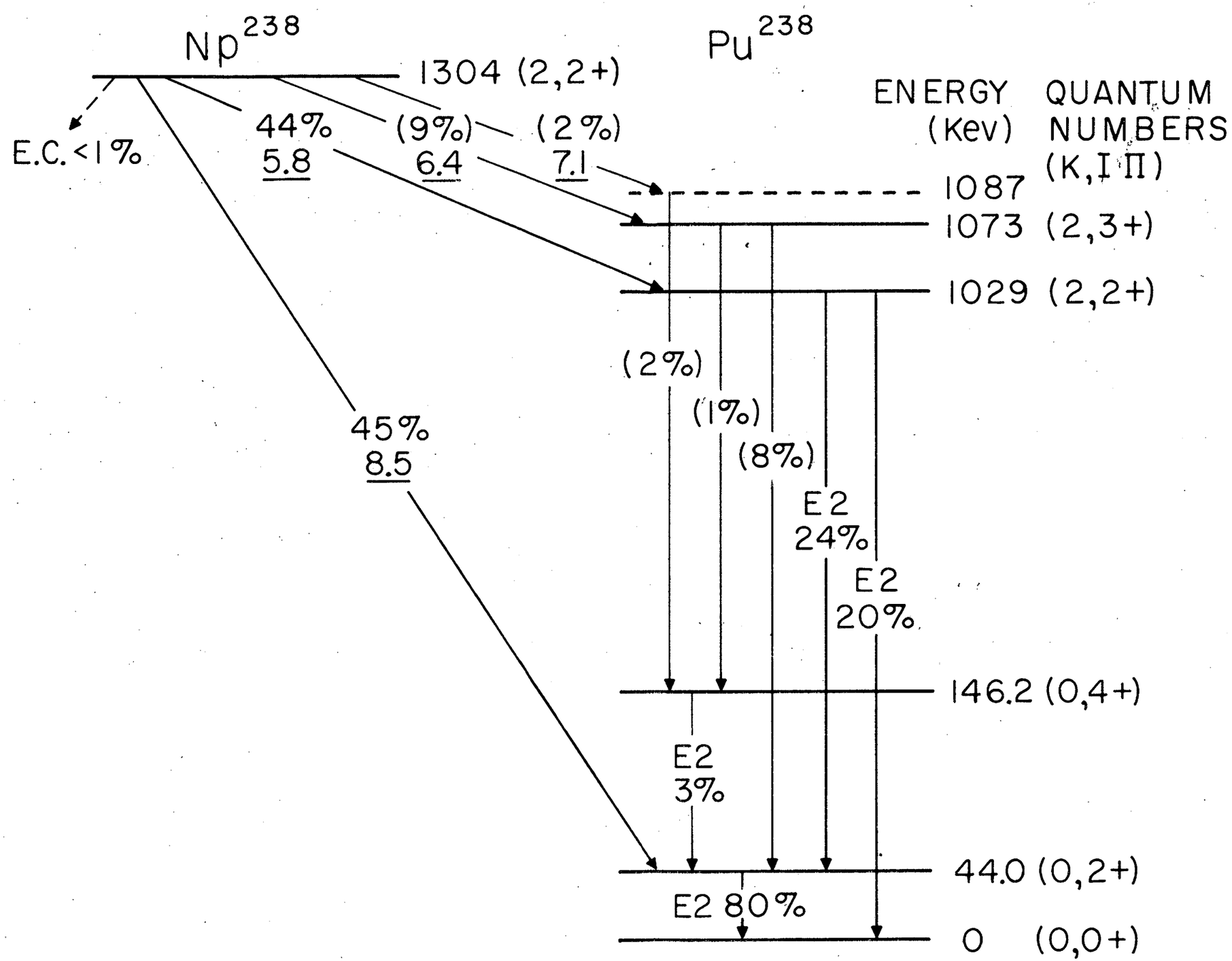

CONVERSION
ELECTRON PEAK
~900 Kev
SOFT BETA
END POINT
~270 Kev
HARD BETA
END POINT
~1260 Kev
COUNTS PER CHANNEL
200
150
100
50
0
10
20
30
40
CHANNEL NUMBER

Fig. 2 23039-2



MU-8617

Fig 3 22845-2

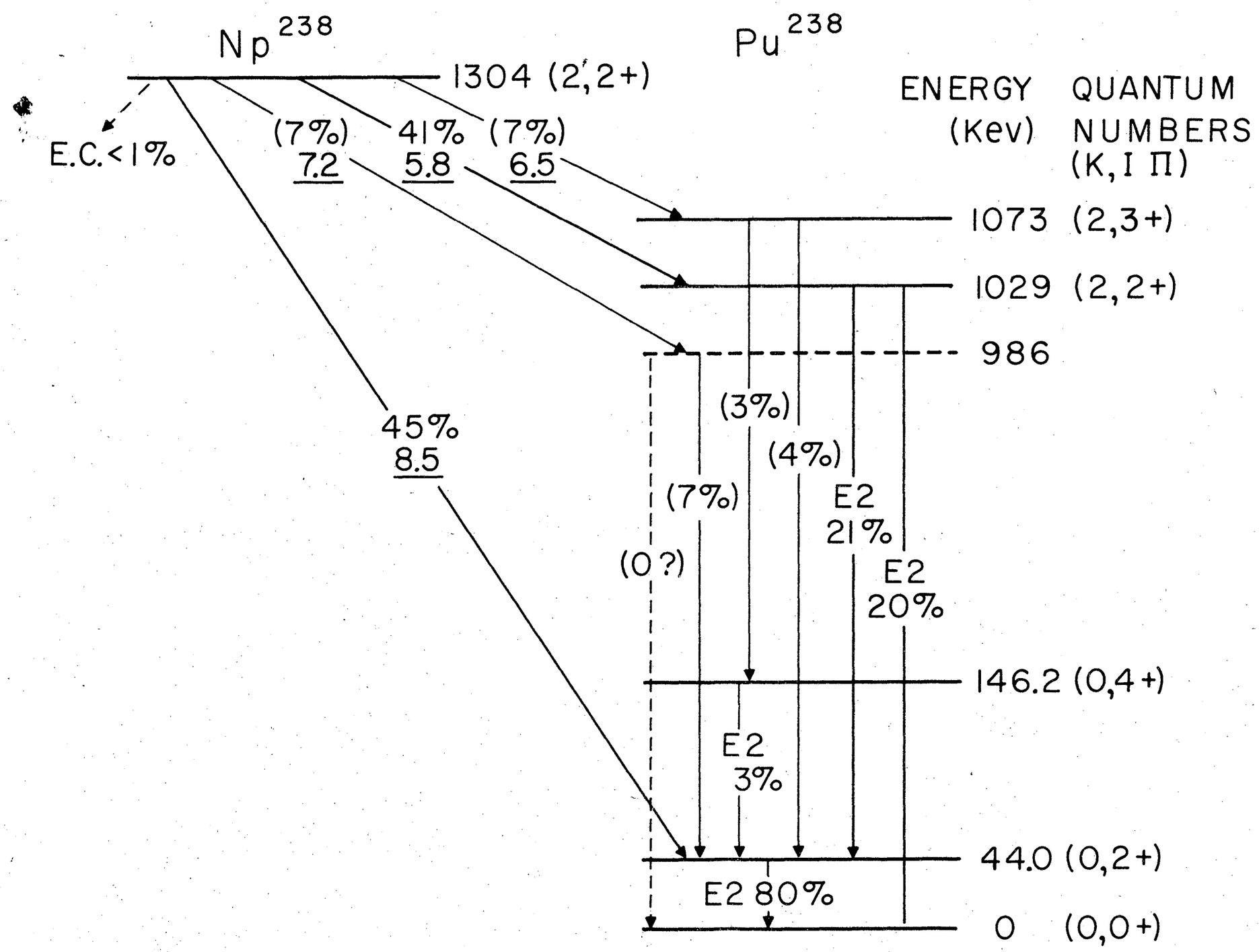

Np 238
Pu 238
1304 (2',2+)
E.C.<1%
(7%)
7.2
41%
5.8
(7%)
6.5
45%
8.5
ENERGY (Kev)
QUANTUM NUMBERS (K, I Π)
1073 (2,3+)
1029 (2,2+)
986
(3%)
(4%)
(7%)
E2
21%
(0 ?)
E2
20%
146.2 (0,4+)
E2
3%
44.0 (0,2+)
E2 80%
0 (0,0+)

Fig 4 23053-2