International Atomic Energy Agency

INDC(CCP)-51/L

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INSTITUTE OF PHYSICS AND POWER ENGINEERING

COMPETITION BETWEEN BETA DECAY AND NEUTRON EMISSION IN ^{237}Np FISSION

B.P. Maksyutenko, Yu.F. Balakshev
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(Obninsk - 1973)

Vienna 1975

IAEA NUCLEAR DATA SECTION, KÄRNTNER RING 11, A-1010 VIENNA

73-9431
Translated from Russian

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UDC 539.173.84

ABSTRACT

From data on the relative yields of delayed neutrons from the pure precursors ^{87}Br, ^{88}Br and ^{137}I in ^{237}Np fission induced by 0.4-1.2 MeV and 3.9-5.1 MeV neutrons, values have been determined for the most probable charge of the light and heavy fragments.

On this basis we have determined the variation with energy of the average number of prompt fission neutrons, this variation being in agreement with that obtained by direct measurement. In addition, the process of energy redistribution between light and heavy fragments and the competition between the two branches of the process - beta decay and neutron emission - are examined.

Study of the variations in the relative yields of delayed neutrons during fission induced by fast neutrons can provide valuable information on the variations in charge distribution, the excitation energy of the fragments and the distribution of that energy between light and heavy fragments, etc. In order to obtain this information, however, it is necessary to know the relative yields of at least some of the pure precursors; only then will it be possible to attempt a physical interpretation of the results of experiments. Hitherto the methods of processing experimental data on the decay in the activity of delayed neutrons did not enable us to understand or solve such problems as the resolution of the exponents in a multicomponent mixture of contributors. In our analysis we separated the only pure group with a half-life of 55.6 sec, which represented the emission of the precursor ^{87}Br. However, this is clearly inadequate for any kind of interpretation of the data.

The new approach to the study of the distribution of the precursors of delayed neutrons [1] permitted a clear formulation of the problem of exponent resolution. It was shown that a set of experimental points representing the decay in the activity of delayed neutrons from a single precursor can be described in terms of the distribution of yield probability as a function of half-life. This is a bell-shaped function which is characterized by three parameters: the position of the maximum on the axis of half-lives, the width and the amplitude. Hence the problem of determining the parameters of the contributors (the half-life and the amplitude) to a multicomponent curve representing the decay in the activity of delayed neutrons reduces to a problem of the spectral Rayleigh type, and this itself can be interpreted as representing the resolution of two or a larger number of exponents. It was shown that the decay in the activity of delayed neutrons is described by a distribution with four maxima (cf. Fig. 1, i.e. where $T_{\frac{1}{2}}$ is approximately 0.2; 2; 20; and 55 sec. Of these, the maximum where $T_{\frac{1}{2}} \approx$ 55 sec corresponds to a single precursor (^{87}Br); the distribution with the maximum at $T_{\frac{1}{2}} \approx 20$ sec corresponds to two main contributors, ^{137}I ($T_{\frac{1}{2}} = 24.7$ sec) and ^{88}Br ($T_{\frac{1}{2}} = 16.3$ sec), with a slight admixture of ^{141}Cs ($T_{\frac{1}{2}} = 24.9$ sec). The second peak has a minimum at $T_{\frac{1}{2}} \approx 8$ sec from the shorter half-lives, and the following maximum at $T_{\frac{1}{2}} \approx 2$ sec corresponds to an undetermined mixture of some ten contributors. On the flank of the peak (in the range from 2 to 8 sec) the large contributors, ^{89}Br (4.45 sec) and ^{138}I (6.3 sec), are represented.

On the basis of further mathematical experiments it was established that a mixture of two exponents shows two distinct maxima (i.e. a separation of these two contributors is achieved) if the interval between their half-lives is approximately 0.4 T, where T is the larger of the two. Even better resolution is found when only the values of the relative yields are sought and the half-lives are given (they are known from radiochemical investigations). In the solution of a simpler problem of this kind the number of parameters sought is substantially reduced (one out of the three remains to be found); moreover, the parameters sought are not contained in the exponent index, so that the accuracy of the result is considerably enhanced.

We shall now show how, on the basis of the yield ratios of groups of delayed neutrons established experimentally, a value can be derived for the most probable charge Z_p of light and heavy fragments, and what conclusions we can draw from it.

The yield of delayed neutrons Y_A from a precursor of mass A, charge Z and emission probability $P_{n,A}$ is determined by the equation

$$Y_A = P(A)P(Z,Z_p)P_{n,A} \tag{1}$$

where P(A) is the probability of the emission of a fragment of mass A, $P(Z,Z_p)$ is the probability of the occurrence of charge Z in the case of the most probable charge Z_p for a given mass A.

For the two isotopes of the fission products of mass A and A + 1 we have from (1):

$$\ln \left[\frac{Y_{A+1}}{Y_A} \cdot \frac{P_{n,A}}{P_{n,A+1}} \cdot \frac{P(A)}{P(A+1)} \right] = \frac{k}{\sigma_A^2} \left[Z - Z_p - \frac{k}{2} \right] \tag{2}$$

where σ_A is the width of the charge distribution and $k = \partial Z_p / \partial A$.

On the basis of (2) Z_p values were calculated for A = 87 by reference to the delayed neutron yields from ^{88}Br and ^{87}Br. In these calculations it was assumed that:

(a) The charge distribution is described by a Gaussian;

(b) The width of the Gaussian and the value of $\partial Z_p / \partial A$ are the same at all the neutron energies studied; and

(c) We are dealing with the direct yield of fragments in fission and not a cumulative yield.

The following parameter values were assumed:

σ_A = 0.59 charge unit

k = 0.486 charge unit/mass unit

$P_{n,A} = 2.4$; $P_{n,A+1} = 4.0$

Data on the mass yields were derived from investigations of the kinetic energies of fission fragments [2] (at an energy lower than 0.8 MeV they were assumed to be the same as at 0.8 MeV in the experiment referred to). Our results for the determination of the relative yields of delayed neutrons are presented in Table 1 and in Figs 2 and 3.

The Zp values were also calculated for a heavy fragment (A = 137), on the basis of equation (1) and the same assumptions, in the formula:

$$Z_{pT} = Z_T + \sqrt{(Z_L - Z_{pL})^2 - 2\sigma_A^2 \ln \frac{Y_T}{Y_L} \cdot \frac{P_{nL}}{P_{nT}} \cdot \frac{P(A_L)}{P(A_T)}} \qquad (3)$$

Here the indices L and T denote light and heavy fragments respectively; in the calculations the following values were adopted:

$Z_T = 53$; $A_T = 137$; $P_{nL} = 2.4$;
$Z_L = 35$; $A_L = 87$; $P_{nT} = 4.8$.

The results are presented in Table 2 and in Fig. 4. The following mode of representation was adopted: the points denote the values $S_L = Z_p(E) - Z_p(0.6)$, while the triangles denote the negative value of S_T. In the upper part of Fig. 4 a straight line has been drawn through the points denoting $S = S_L + S_T$ (the calculation was carried out by the method of least squares).

Explanations of the tables will be presented later, but now we shall turn to an analysis of the results.

Results

As was shown in Ref. [3], the rate of change in the average number of prompt fission neutrons is associated with the rate of change in Zp as follows:

$$\frac{\partial \bar{\nu}}{\partial E} = \frac{1}{k} \frac{\partial S}{\partial E}$$

where $S = S_L + S_T$.

By applying the method of least squares to the points representing $S = S_L + S_T$, we found the value of $\partial S/\partial E$, and from it:

$$\partial \bar{\nu}/\partial E = 0.148 \pm 0.032 \ \frac{\text{neutrons}}{\text{MeV}} ,$$

which is in excellent agreement with the results of direct measurements of this value and confirms the validity of our result.

Thus, the value of the most probable total fragment charge increases with the energy of the fission-inducing neutrons at a rate proportional to the increase in the number of prompt fission neutrons; the fragments become less overloaded with neutrons and approach the line representing stable nuclei.

We shall now consider how this process of variation in charge is reflected in the light and heavy fragments. We have two sets of data (0.4-1.2 MeV and 3.9-5.1 MeV) divided by an interval in which these investigations were not yet carried out. We shall mention two effects: the cumulative effect, comprising the reduction (increase) of the most probable value of the light (heavy) fragment charge on transition from one region to another, and a local effect, comprising a smooth variation represented by a bell-shaped curve. We shall regard the excitation energy of a fragment as consisting of two parts: (a) the energy distributed over all nucleons, so that it can be produced only as a result of a structural change in the nuclear fragment, i.e. as a result of beta decay, and (b) the energy which can become concentrated on one of the neutrons, so that its emission becomes possible. Thus for a light fragment the process can be described as follows: in the range from 1.2-3.9 MeV a reduction in Z_p (points $1 \rightarrow 2$ in Fig. 5) signifies an increase in the length of the beta decay chain; in this case the fragment moves away from the Z_A stability line and the share of the excitation energy distributed over all its nucleons increases. Ultimately the moment comes when this energy becomes quite substantial and is concentrated on one of the neutrons, which is then emitted by a light fragment (transition 2 → 3 in Fig. 5, region E_n0:3.9-5.1 MeV). Actually, it is impossible to speak of smooth variation in the length of the beta decay chain in relation to some individual fragment. In the case of individual fragments the chain length cannot be expressed by a fraction. A smooth variation in the beta decay chain length is the result of averaging over many nuclear fragments. This fact should therefore be interpreted as meaning that only a small, but steadily increasing, portion of nuclear fragments can concentrate energy on a single neutron so that the latter is emitted, while the vast majority of them undergo beta decay. When we approach an energy $E_p = 4.8$ MeV this probability of energy concentration of a single nucleon is so high that the process is already in the nature of a fluctuation for all fragments and the neutron emission becomes preferential.

The nature of the variation in the excitation energy in a heavy fragment is exactly the opposite in nature: in the vicinity of $E_p = 4.8$ MeV there is a reduction in the excitation energy and a transfer of that energy to a light fragment with subsequent emission of the neutron by it.

At the threshold of ^{237}Np fission there is a reduction in the excitation energy of a light fragment and an increase in that of a heavy fragment, but without the additional emission of a neutron. It is evident that such a method of fission is more favourable from the energy point of view.

We should add that the sum of the most probable charges of the additional (by mass) light and heavy fragments in the case of ^{237}Np is in complete agreement with similar data on ^{235}U (n_{th},f). The charge Z_p (total) for these fragments can be found as follows:

Z_p (total) = $Z_p(87) + Z_p(137) + k(A_F - \bar{\nu} - 87 - 137)$, where A_F is the mass number of the fissile compound nucleon. The results were as follows:

For ^{235}U (n_{th},f) Z_p (total) = 92.45 charge units

For ^{237}Np (1.1 MeV) Z_p (total) = 93.46 charge units

The difference between these numbers is equal to the difference between the charges of the fissile nuclei.

In order not to confuse the main subject matter of this article we have postponed the discussion of some details to the following explanatory paragraphs.

1. The technical details of the experiment have been described previously, and here we shall merely recall that in the energy range from 0.4-1.2 MeV the I(p,n)^{3}He reaction was used to obtain neutrons, the target thickness having been approximately 0.9 mg/cm^2, and in the region from 3.9-5.1 MeV the D(d,n)^{4}He reaction was used, with a target thickness of approximately 1.5 mg/cm^2.

2. A new method [1] was used to determine the yields of ten groups of delayed neutrons. For the first three the half-lives of the pure precursors ^{87}Br, ^{137}I and ^{88}Br were given. The half-life of the following group, which comprised a mixture of three precursors with a comparable contribution and similar half-lives (6.3 sec, ^{138}I, 5.9 sec, ^{93}Rb and 5.8 sec, ^{87}Se) was given by some average value (6.1 sec). The yield of the following group, which represented an almost pure contribution from the precursor ^{89}Br, was determined from its half-life. Where there was no longer any expectation of splitting, i.e. in the case of any mixture of short-lived groups, we found a distribution with uniform spacing of 0.5 sec in the 0-2.5 sec range along the half-life axis. Calculations showed that a resolution of 6.1 and 4.45 sec for the group are almost never achieved although, as can be seen from the figures, the relative yield of the fourth group, determined largely for ^{138}I, doubles the yield of ^{137}I, and the yield of the "purer" group (4.45 sec) does not quite reproduce the variation in the ^{88}Br/^{87}Br yield. The reason is probably the effect of the larger contributors on the left-hand and right-hand sides. In the 3.9-5.1 MeV range the yield values are an average derived from the separate processing of 2-4 sets of measurements, each containing the sum of 20-30 decay curves. The mean square error was determined on the basis of their scatter. In the lower energy region a composite decay curve (covering all sets of measurements) was prepared for each energy: in these an error was assigned to each result in accordance with the statistical accuracy and the average relative error in the upper region.

3. The contribution derived as a result of ^{87}Se → ^{87}Br beta decay in relation to the direct yield from ^{87}Br, like the contribution from the beta decay of ^{88}Se → ^{88}Br when $E_p = 1.2$ MeV is a few per cent, and the calculation of Z_p carried out was found to be sufficiently correct. At an energy of 0.9 MeV this contribution becomes substantial, similar in order of magnitude to that for the fission of ^{235}U (n_{th},f). For this case we calculated Z_p by another method, using data relating to the fission of ^{235}U (n_{th},f) as a reference. The relationship takes the form:

$$Z_{pb} - Z_{pa} = - \frac{\sigma_A{}^2}{k} \ln \left[\frac{Y_{A+1}}{Y_A} \frac{P(A)}{P(A+1)} \right]_b \left[\frac{Y_A}{Y_{A+1}} \frac{P(A+1)}{P(A)} \right]_a, \qquad (4)$$

where the subscript "a" relates to $^{235}U\ (n_{th}, f)$ and the subscript "b" relates to ^{237}Np. In equation (4), as opposed to equation (2), we exclude the not very exactly known values of Pn, which naturally improves the result, but we introduce the additional error of the reference values. Calculations based on (2) and (4) differ by 0.1 charge unit.

4. The reason for the large scatter of Zp values when $E_p = 1.2$ MeV is still unclear to us. In the calculation of $\partial\bar{\nu}/\partial E$ this point was not used.

REFERENCES

[1] TARASKO, M.Z., MAKSYUTENKO, B.P., Paper 369 Instit. of Phys. and Power Eng. (1972).

[2] KUZMINOV, B.D., et al., Jadernaja fizika 11 (1970) 297.

[3] MAKSYUTENKO, B.P., Jadernaja fizika 17 (1973) 481.

Table 1

Relative yields of delayed neutrons in ^{237}Np fission

Group No.	$T_{\frac{1}{2}}$ (sec)	Relative yields (Y_i/Y_1) E_p = 0.4 MeV	E_p = 0.6 MeV
1	55,6	1	1
2	24,7	6,87 ± 0,82	8,13 ± 0,41
3	16,3	2,33 ± 0,42	1,55 ± 0,18
4	6,1	4,8 ± 1,4	6,28 ± 0,69
5	4,45	3,91 ± 0,55	4,23 ± 0,51
6	2,5	9,44 ± 0,66	7,37 ± 0,59
7	2,0	6,73 ± 0,67	6,59 ± 0,52
8	1,5	1,80 ± 0,36	3,03 ± 0,55
9	1,0	0,197 ± 0,054	0,67 ± 0,12
10	0,5	0,068 ± 0,021	0,31 ± 0,08

Group No.	$T_{\frac{1}{2}}$ (sec)	Relative yields (Y_i/Y_1)		
		E_p = 0.7 MeV	E_p = 0.8 MeV	E_p = 0.9 MeV
1	55,6	1	1	1
2	24,7	6,90 ± 0,34	6,83 ± 0,20	6,47 ± 0,19
3	16,3	2,07 ± 0,18	2,21 ± 0,21	2,55 ± 0,18
4	6,1	3,55 ± 0,35	3,31 ± 0,30	3,34 ± 0,33
5	4,45	5,29 ± 0,63	5,55 ± 0,72	6,84 ± 0,75
6	2,5	7,35 ± 0,59	7,71 ± 0,69	7,22 ± 0,36
7	2,0	5,37 ± 0,43	5,01 ± 0,35	4,29 ± 0,69
8	1,5	2,46 ± 0,39	2,00 ± 0,34	1,85 ± 0,26
9	1,0	0,72 ± 0,13	0,56 ± 0,08	0,83 ± 0,15
10	0,5	0,49 ± 0,10	0,37 ± 0,08	0,52 ± 0,11

Group No.	$T_{\frac{1}{2}}$ (sec)	Relative yields (Y_i/Y_1)		
		E_p 1.0 MeV	1.1 MeV	1.2 MeV
1	55,6	1	1	1
2	24,7	6,73 ± 0,17	7,10 ± 0,17	8,11 ± 0,23
3	16,3	2,00 ± 0,16	1,89 ± 0,19	1,07 ± 0,13
4	6,1	4,01 ± 0,36	5,30 ± 0,52	6,89 ± 0,76
5	4,45	5,57 ± 0,72	3,82 ± 0,52	5,00 ± 0,70
6	2,5	7,19 ± 0,36	8,54 ± 0,34	7,91 ± 0,32
7	2,0	4,46 ± 0,31	7,46 ± 0,52	6,70 ± 0,40
8	1,5	1,74 ± 0,28	2,79 ± 0,36	3,23 ± 0,38
9	1,0	0,60 ± 0,10	0,40 ± 0,06	0,94 ± 0,13
10	0,5	0.60 ± 0,21	0,15 ± 0,03	0,66 ± 0,12

Group No.	$T_{\frac{1}{2}}$ (sec)	Relative yields (Y_i/Y_1)		
		E_p (MeV) 3.9	4.2	4.5
1	55,6	1	1	1
2	24,7	4,29 ± 0,14	4,322 ± 0,052	4,537 ± 0,035
3	16,3	2,28 ± 0,13	2,06 ± 0,17	1,665 ± 0,094
4	6,1	2,70 ± 0,30	2,74 ± 0,28	3,81 ± 0,30
5	4,45	5,21 ± 0,09	5,42 ± 0,52	4,44 ± 0,26
6	2,5	9,86 ± 0,50	9,98 ± 0,66	8,63 ± 0,67
7	2,0	6,244 ± 0,028	5,75 ± 0,27	6,72 ± 0,56
8	1,5	2,13 ± 0,14	2,01 ± 0,33	2,69 ± 0,48
9	1,0	0,436 ± 0,051	0,60 ± 0,14	0,512 ± 0,061
10	0,5	0,233 ± 0,059	0,44 ± 0,10	0,297 ± 0,095

Group No.	$T_{\frac{1}{2}}$ (sec)	Relative yields (Y_i/Y_1)	
	E_p (MeV)	4.8	5.1
1	55,6	1	1
2	24,7	4,93 ± 0,26	4,18 ± 0,48
3	16,3	1,51 ± 0,22	2,10 ± 0,38
4	6,1	4,40 ± 0,59	3,04 ± 0,83
5	4,45	5,17 ± 0,33	4,84 ± 0,65
6	2,5	9,36 ± 0,45	10,05 ± 0,47
7	2,0	7,53 ± 0,64	7,0 ± 1,0
8	1,5	3,46 ± 0,45	2,40 ± 0,62
9	1,0	0,839 ± 0,069	0,376 ± 0,061
10	0,5	0,374 ± 0,025	0,164 ± 0,023

Table 2

Values of Z_p for A = 87 and A = 137 and the sum of these values

E_p (MeV)	Z_p		
	A = 87	A = 137	Total
0,4	34,70	53,41	88,11
0,5			
0,6	34,97	53,18	88,15
0,7	34,76	53,37	88,13
0,8	34,72	53,41	88,13
0,9	34,62	53,53	88,15
1,0	34,81	53,40	88,21
1,1	34,86	53,35	88,21
1,2	35,26	53,28	88,54
3,9	34,70	53,66	88,36
4,2	34,78	53,62	88,40
4,5	34,93	53,55	88,48
4,8	35,00	53,48	88,48
5,1	34,75	53,64	88,39

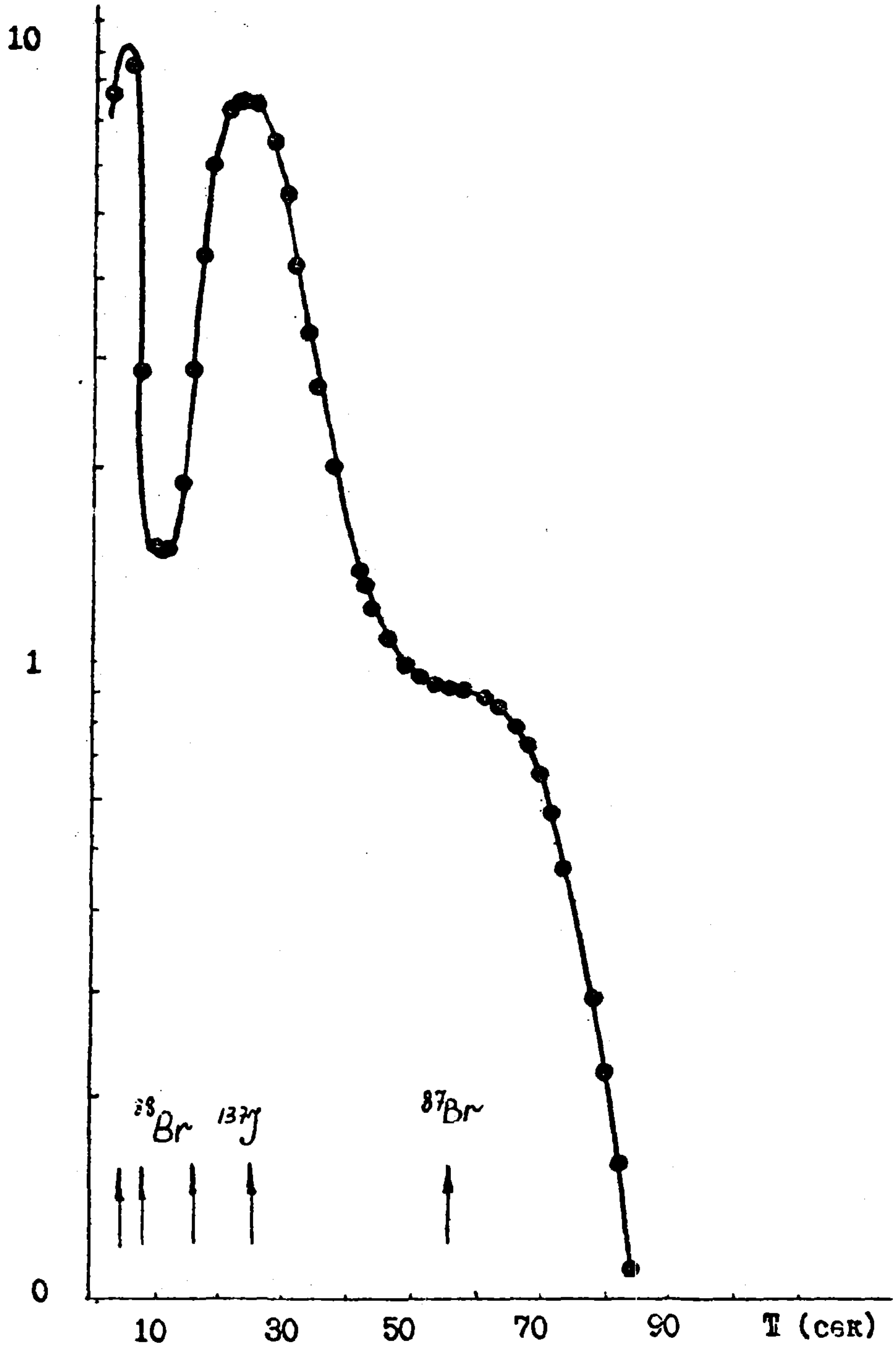

Fig. 1 Distribution of the probability of delayed neutron emission as a function of half-life in the case of ^{237}Np.

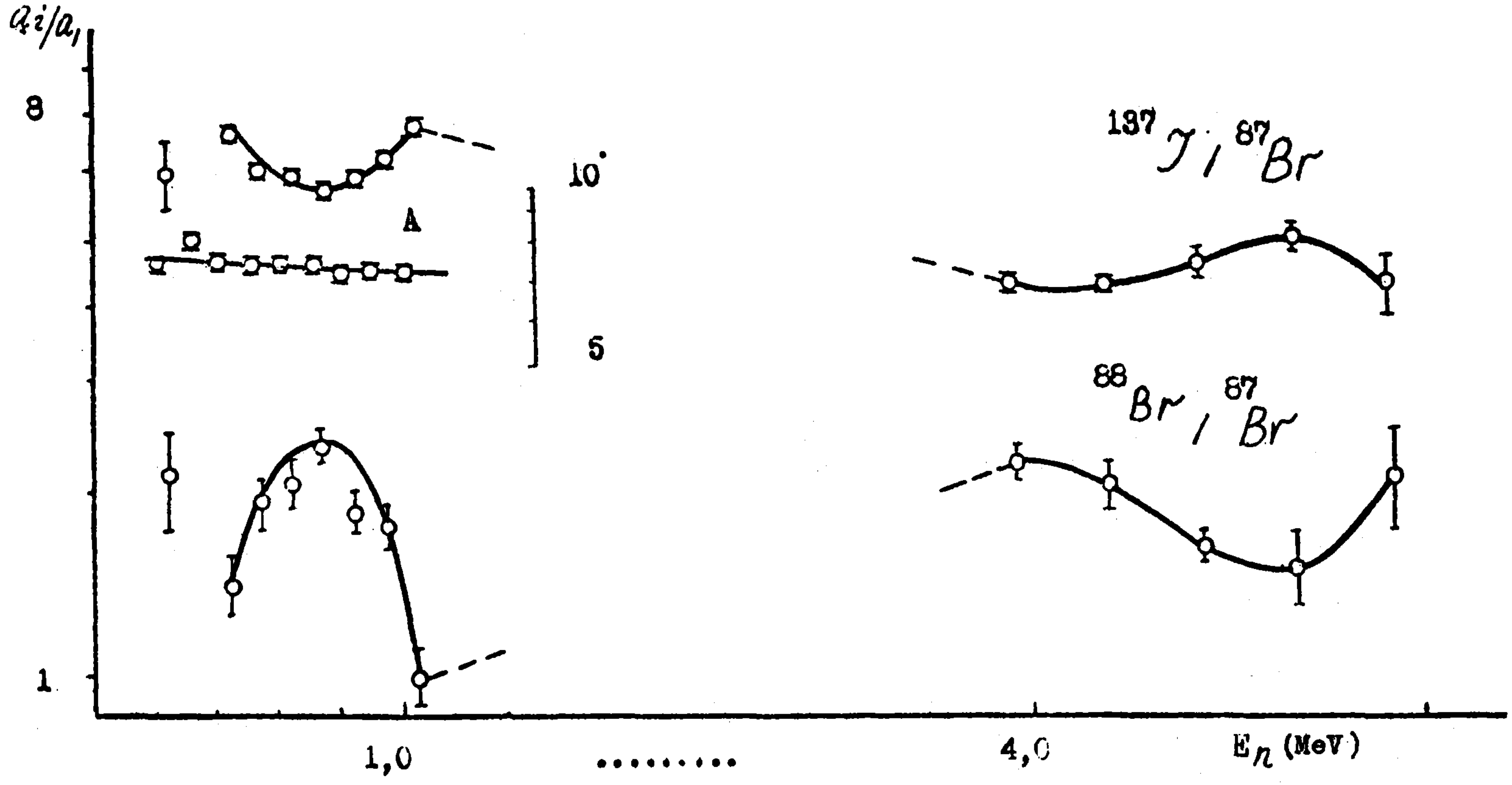

Fig. 2 Relative yields of delayed neutrons from ^{137}I and ^{88}Br.

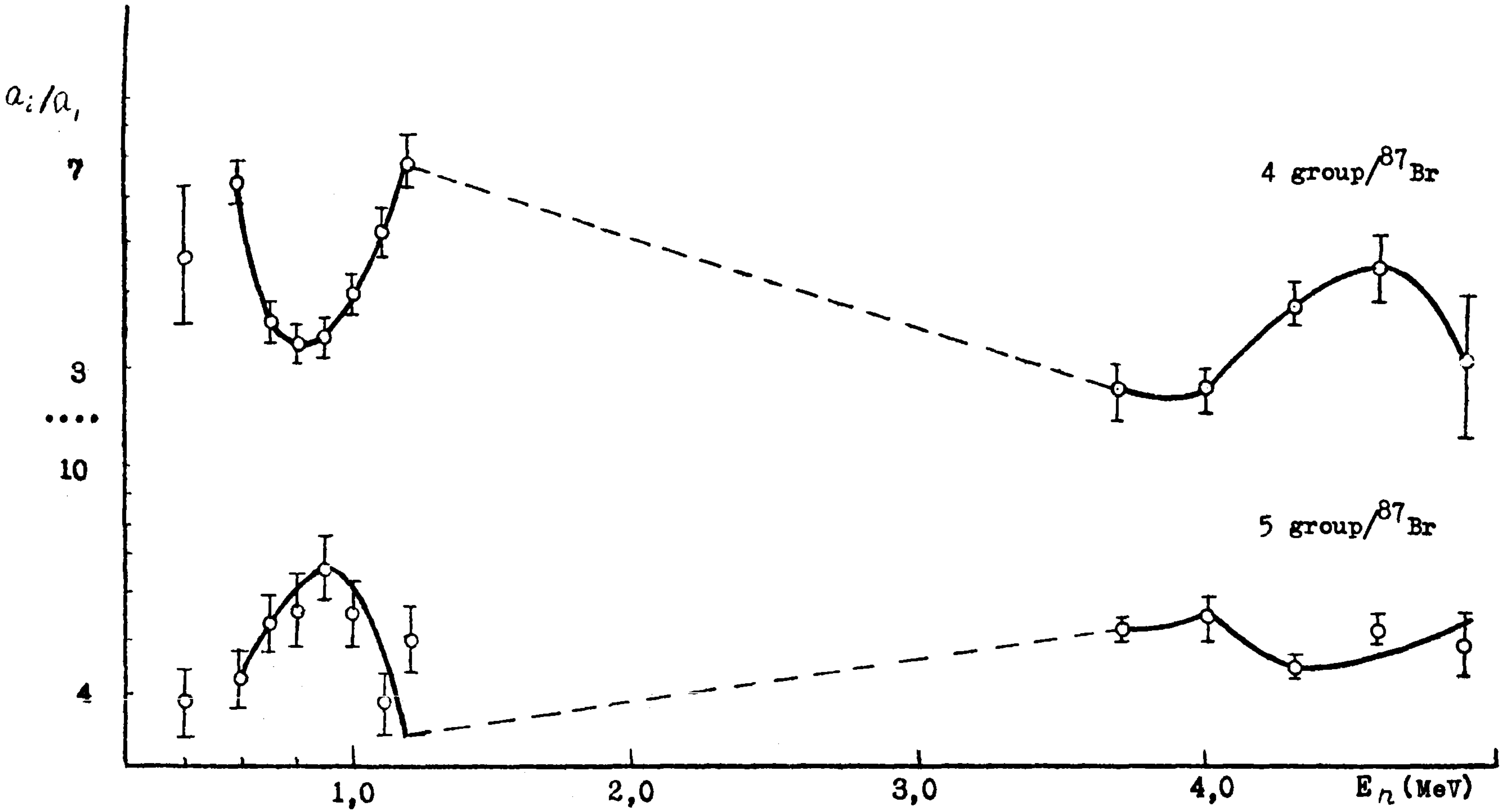

Fig. 3 Relative yields of delayed neutrons of the fourth and fifth groups.

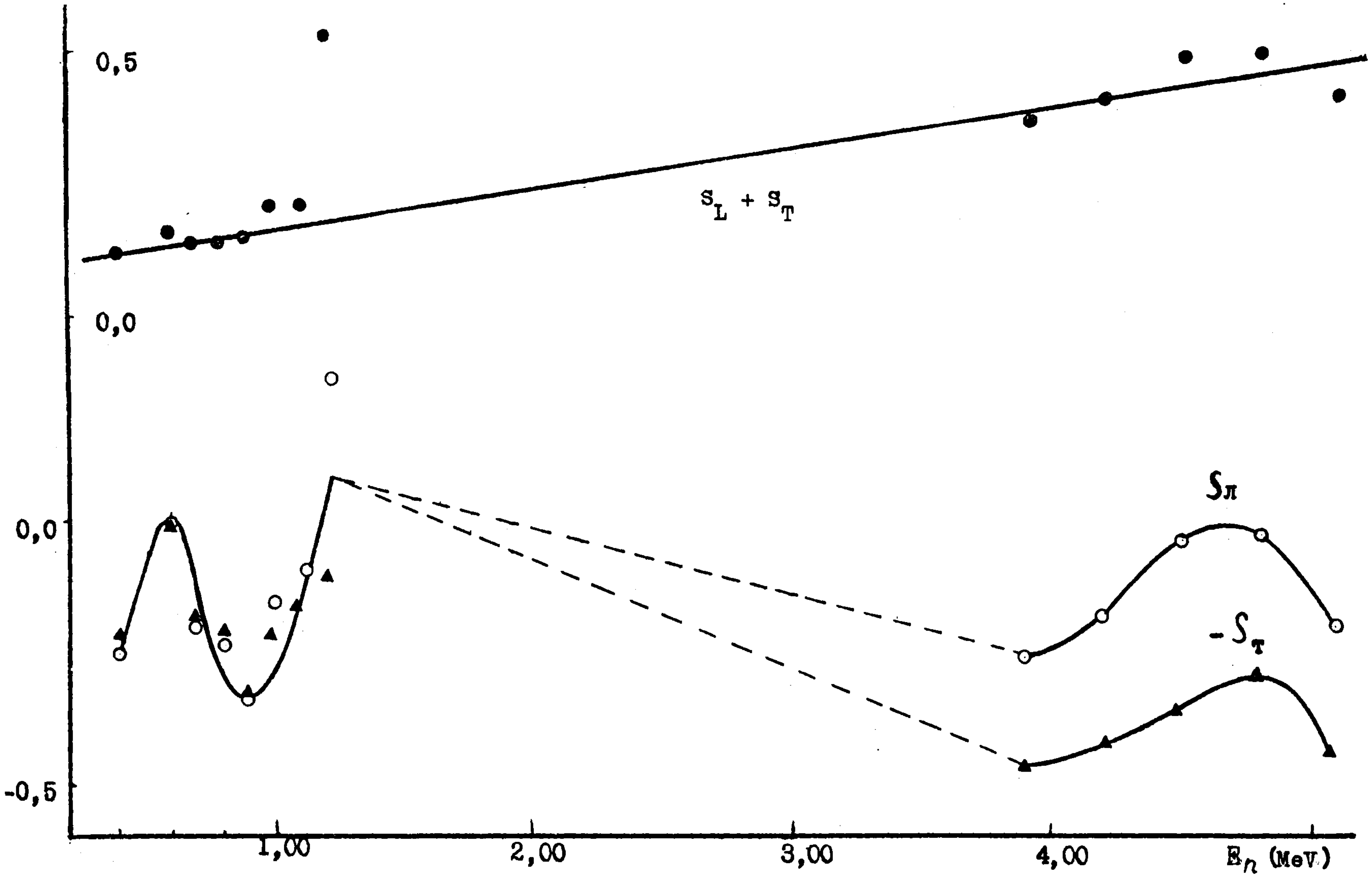

Fig. 4 Variation in the most probable charge.

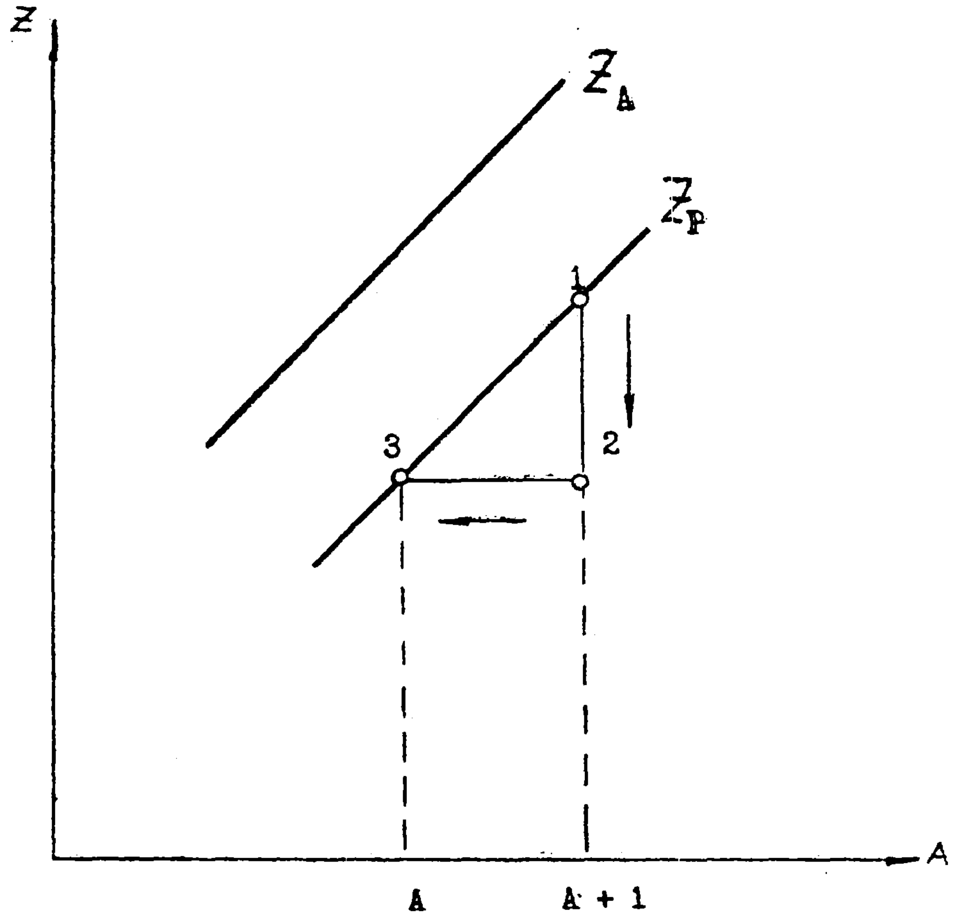

Fig. 5 Diagram of neutron and β radiation.