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Delayed-neutron activities produced in fission: Mass range 122–146

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Delayed-neutron emission from mass separated heavy fission products has been studied using a sensitive neutron counter. Twelve new delayed-neutron precursors have been found, namely ^{122}Ag, ^{123}Ag, ^{127}In, ^{128}Cd (or ^{128}In^m), ^{128}In, ^{129}In (two isomers), ^{130}In, ^{131}In, ^{132}In, ^{133}Sn, and ^{136}Sb. The half-life determination for 11 other precursors has been improved.

[RADIOACTIVITY ^{122,123}Ag, $^{127-132}$In, ^{128}Cd, ^{133,134}Sn, $^{134-136}$Sb, ^{136}Te, $^{137-141}$I, $^{141-146}$Cs, measured delayed neutrons; deduced half-lives.]

I. INTRODUCTION

The experimental technique used in the half-life determinations of mass-separated delayed-neutron precursors produced in fission has been outlined in the first part of the present study.[1] Accurate half-life determinations of 20 precursors in the mass range 79–98, 5 of them new, were then given. In the present part of the study the mass range 120–147 has been covered yielding half-lives of 12 new delayed-neutron precursors and redeterminations in 15 other cases.

II. EXPERIMENTAL RESULTS

The experimental results are collected in Table I. The errors of the individual measurements (fourth column) correspond to one standard deviation based on the fit of the experimental points to the calculated decay curve. These errors are purely statistical. The error of the average value (fifth column) is one standard deviation computed from the reproducibility of the measurements. The average value is a weighted mean value unless the errors of the different determinations are inconsistent with the reproducibility (too small). Then, these errors are disregarded, and the average value is unweighted.

When the internal and external consistencies have indicated an error below 0.5% this error has somewhat arbitrarily been increased to 0.5%. For nuclides with only one determination relative errors smaller than 2% have been arbitrarily adjusted to this value. In order to acquire sufficient statistics most cases have been repeated 50–200 times using an automatic multiscaling technique (cf. Ref. 1).

All nuclides have also been measured with a container with heavy water placed close to the source in order to check the effect of photoproduced neutrons. The half-lives obtained in this way are not included in the determination of the average values although the results were in agreement with measurements without heavy water. The heavy water effect was found to be negligible in all cases except for mass 131 where photoneutrons may account for about 10% of the neutrons.

Table II lists the relative "neutron windows" $(Q_\beta - B_n)/Q_\beta$ calculated from four current mass formulas[2-5] and from experimental or extrapolated values taken from the 1971 Mass Table by Wapstra and Gove.[6]

Decay curves for some typical cases are shown in Figs. 1–3. The decay has normally been followed longer than indicated in the figures. The errors (± one standard deviation) are shown unless they are smaller than the size of the circles. It should be noted that in most cases only part of the measured points have been plotted.

A. Mass number 122

A weak activity has been found at mass number 122 with a half-life of about 1 sec. From Table II it is found that ^{122}Cd is not likely to be a delayed-neutron precursor while ^{122}Ag has a positive neutron window. Moreover, the half-life is in agreement with the value 1.5 ± 0.5 sec attributed to ^{122}Ag and obtained from γ-ray studies at this laboratory.[7] Therefore we conclude that the weak activity at mass 122 is due to the new neutron precursor ^{122}Ag.

B. Mass number 123

From mass formula predictions the indium and cadmium isobars of this mass are not likely to be delayed-neutron precursors. Silver, on the other hand, has a positive neutron window. Consequently, it seems probable that the 0.39 sec activity is due to ^{123}Ag.

TABLE I. Experimental results on delayed-neutron precursors.

Mass number	Element assignment	Collection time (sec)	Half-life (sec)	Half-life (sec) Average value this work	Half-life (sec) From β decay (Ref. 8)	Half-life (sec) Literature
123	Ag	2	0.39 ± 0.03	0.39 ± 0.03		
127	In	1	3.63 ± 0.06			
		5	3.77 ± 0.02	3.76 ± 0.03	3.7 ± 0.1	
128	In (Cd)	2	0.94 ± 0.06			
		30	1.07 ± 0.10			
		5	0.75 ± 0.05			
		2	0.96 ± 0.02	0.94 ± 0.05	0.80 ± 0.03	
	In	10	16.1 ± 0.5			
		30	12.0 ± 0.2			
		5	9.2 ± 0.8		5.6 ± 0.4	
129	In	1	0.98 ± 0.05			
		2	0.99 ± 0.02			
		4	1.01 ± 0.04			
		1	0.98 ± 0.04	0.99 ± 0.02	0.8 ± 0.3	
	In	2	2.49 ± 0.10			
		4	2.32 ± 0.29			
		1	2.2 ± 0.4	2.5 ± 0.2		
130	In	2	0.576 ± 0.10	0.58 ± 0.01		0.53 ± 0.05 [a]
131	In	4	0.277 ± 0.004			
		1	0.283 ± 0.009			
		60	0.302 ± 0.005	0.29 ± 0.01		0.27 ± 0.012 [b]
132	In	1	0.25 ± 0.03			
		1	0.45 ± 0.07	0.3 ± 0.1		0.12 ± 0.02 [c]
133	Sn	2	1.52 ± 0.12			
		2	1.44 ± 0.09	1.47 ± 0.07		1.47 ± 0.04 [d]
134	Sn	1	1.009 ± 0.010			
		2	1.07 ± 0.03			
		10	1.053 ± 0.012			
		20	1.15 ± 0.04	1.04 ± 0.02		0.7 ± 0.2 [e]
	Sb	10	10.7 ± 0.2			11.3 ± 0.3 [f]
		20	9.92 ± 0.11	10.3 ± 0.4	10.2 ± 0.3	11.1 ± 0.8 [g]
						10.3 ± 0.15 [h]
135	Sb	4	1.819 ± 0.016	1.82 ± 0.04		1.70 ± 0.02 [i]
136	Sb	1	0.823 ± 0.013			
		2	0.93 ± 0.15	0.82 ± 0.02		
	Te	25	17.51 ± 0.04	17.5 ± 0.4		20.9 ± 0.5 [j]
						24 ± 2 [k]
137	I	6	23.89 ± 0.31			
		30	24.25 ± 0.03	24.25 ± 0.12	24.5 ± 0.2	24.62 ± 0.08 [l]
138	I	2	6.61 ± 0.02			
		8	6.31 ± 0.04	6.46 ± 0.15	6.62 ± 0.09	6.55 ± 0.11 [l]
139	I	4	2.303 ± 0.009	2.30 ± 0.05	2.47 ± 0.15	2.61 ± 0.11 [l]
140	I	2	0.585 ± 0.003			0.89 ± 0.12 [m]
		1	0.602 ± 0.004	0.59 ± 0.01		0.86 ± 0.04 [m]
141	Cs	30	22.15 ± 0.19	22.2 ± 0.4	25.6 ± 0.3	24.9 ± 0.2 [n]
						24.7 ± 0.4 [o]
	I	3	0.513 ± 0.007			
		1	0.449 ± 0.003	0.48 ± 0.03		0.41 ± 0.08 [m]
142	Cs	4	1.79 ± 0.03			1.94 ± 0.01 [o]
		2	1.599 ± 0.012	1.69 ± 0.09		1.68 ± 0.02 [n]
143	Cs	2	1.779 ± 0.008			1.60 ± 0.14 [p]
		2	1.779 ± 0.003	1.78 ± 0.01		1.69 ± 0.13 [q]
						1.7 ± 0.1 [r]

TABLE I *(Continued)*

Mass number	Element assignment	Collection time (sec)	Half-life (sec)	Half-life (sec) Average value this work	From β decay (Ref. 8)	Literature
144	Cs	1.5	1.02 ± 0.01			1.05 ± 0.14 [q]
		1.5	0.980 ± 0.003	1.00 ± 0.02		1.06 ± 0.10 [p]
145	Cs	1	0.584 ± 0.002	0.58 ± 0.01		0.563 ± 0.027 [s]
						0.611 ± 0.021 [t]
146	Cs	1	0.343 ± 0.007	0.343 ± 0.007		0.189 ± 0.011 [t]
						0.352 ± 0.042 [t]

[a] Reference 12.
[b] Reference 13.
[c] Reference 14.
[d] Reference 15.
[e] Reference 19.
[f] Reference 20.
[g] Reference 21.
[h] Reference 16.
[i] Reference 22.
[j] Reference 23.
[k] Reference 8.
[l] Recommended value from Ref. 24.
[m] Reference 25.
[n] Reference 26.
[o] Reference 27.
[p] Reference 29.
[q] Reference 30.
[r] Reference 31.
[s] Reference 32.
[t] Reference 33.

C. Mass number 127

β counting reveals two short-lived components at this mass number with half-lives 1.3 ± 0.2 and 3.7 ± 0.1 sec,[8] respectively. Experiments with rapid chemical separation have shown that a short-lived Cd activity is present at this mass number.[9] From γ spectroscopy studies, however, two isomers of ^{127}In have been found with half-lives of 1.2 and 3.4 sec, respectively.[10] The longer half-life agrees well with the present determination of 3.76 ± 0.03 sec. In this case one might expect delayed neutrons from indium whereas cadmium is a more doubtful case as seen from Table II.

D. Mass number 128

At this mass number two neutron activities have been found with half-lives 0.94 ± 0.05 and ~12 sec. The former half-life agrees fairly well with the value 0.80 ± 0.03 sec found by β counting, but the latter result has been impossible to reproduce and is at variance with the β counting value 5.6 ± 0.4 sec.[8] The only explanation seems to be that these half-lives correspond to different nuclides.

It seems unlikely that ^{128}Cd should have a half-life as long as 12 sec, and therefore this probably corresponds to an isomer of ^{128}In. The short-lived activity might well be due to ^{128}Cd, especially since rapid chemistry has proven the presence of short-lived cadmium at mass 128 [9]: The assignment of the 0.94 sec activity to another isomer of indium cannot be ruled out, however.

E. Mass number 129

Again two neutron activities have been found with half-lives 0.99 ± 0.02 and 2.5 ± 0.2 sec, respectively. The former half-life agrees with the value 0.8 ± 0.3 sec found by β counting and ascribed to ^{129}In.[11] Since no short-lived cadmium activity has been found at this mass[9] we conclude that both activities are due to isomers of indium.

F. Mass number 130

The single neutron activity of half-life 0.58 ± 0.01 sec found at this mass is presumably due to indium, for which the half-life has been determined to 0.53 ± 0.05 sec by β counting.[12]

G. Mass number 131

The 0.29 sec activity is to be attributed to ^{131}In for which the β half-life has been determined to be 0.27 ± 0.02 sec.[13]

H. Mass number 132

The neutron half-life 0.3 ± 0.1 sec obtained at this mass number is higher than the value 0.12 ± 0.02 sec determined by β counting, and assigned to ^{132}In.[14] Still, it seems probable that the precursor is ^{132}In. The low precision in the present measurement indicates the desirability of further studies. It might be pointed out that an experiment with heavy water present gave the half-life 0.32 ± 0.04 sec, i.e. again higher than the published value.

TABLE II. Relative "neutron window" calculated from various mass formulas.

Nuclide	$(Q_\beta - B_n)/Q_\beta$ from Garvey *et al.* (Ref. 2)	Myers and Swiatecki (Ref. 3)	Seeger (Ref. 4)	Zeldes *et al.* (Ref. 5)	Wapstra and Gove (Ref. 6)
^{120}Ag	0.04	0.03	0.03	0.00	
^{121}Ag	0.19	0.15	0.18	0.13	
^{122}Ag	0.23	0.19	0.15	0.14	
^{122}Cd	−1.11	−0.40	−0.84	−1.23	
^{123}Ag	0.30	0.32	0.29	0.28	
^{123}Cd	−0.43	−0.28	−0.35	−0.46	
^{123}In	−0.34	−0.38	−0.39	−0.33	−0.34
^{124}Ag	0.35	0.30	0.27	0.24	
^{124}Cd	−0.47	−0.22	−0.30	−0.36	
^{124}In	−0.16	−0.14	−0.14	−0.17	−0.13
^{125}Ag	0.40	0.44	0.40	0.38	
^{125}Cd	−0.05	−0.04	−0.06	−0.23	
^{125}In	−0.05	−0.11	−0.07	−0.07	
^{126}Cd	−0.17	0.08	0.00	−0.22	
^{126}In	0.00	0.05	0.03	−0.01	
^{127}Cd	−0.01	0.14	0.10	−0.06	
^{127}In	0.14	0.16	0.15	0.13	
^{128}Cd	0.05	0.27	0.22	0.00	
^{128}In	0.13	0.19	0.19	0.11	
^{129}Cd	0.11	0.27	0.23	0.08	
^{129}In	0.28	0.32	0.29	0.25	
^{130}In	0.24	0.30	0.30	0.21	
^{131}In	0.40	0.43	0.38	0.32	
^{131}Sn	−0.58	−0.35	−0.55	−0.43	
^{132}In	0.43	0.41	0.42		
^{132}Sn	−0.69	−0.30	−0.90	−0.87	
^{132}Sb	−0.31	−0.19	−0.27	−0.31	−0.45
^{133}In	0.71	0.78	0.78		
^{133}Sn	0.01	0.17	0.04	−0.04	
^{133}Sb	−0.21	−0.14	−0.37	−0.26	−0.49
^{134}Sn	0.43	0.59	0.54	0.45	
^{134}Sb	0.15	0.22	0.15	0.13	
^{135}Sb	0.49	0.51	0.57	0.54	
^{135}Te	−0.37	−0.17	−0.26	−0.46	−0.32
^{136}Sb	0.45	0.52	0.48	0.49	
^{136}Te	0.11	0.30	0.26	0.07	
^{136}I	−0.13	−0.07	−0.20	−0.15	−0.14
^{137}Sb	0.58	0.67	0.62	0.65	
^{137}Te	0.16	0.29	0.24	0.15	
^{137}I	0.22	0.36	0.30	0.31	0.28
^{138}Te	0.41	0.46	0.39	0.33	
^{138}I	0.26	0.33	0.26	0.30	
^{139}I	0.42	0.49	0.40	0.46	
^{139}Xe	−0.26	−0.07	−0.17	−0.25	−0.25
^{140}I	0.40	0.43	0.38	0.39	
^{140}Xe	−0.17	0.08	−0.05	−0.18	−0.12
^{140}Cs	−0.02	0.03	−0.10	−0.03	−0.12
^{141}I	0.53	0.58	0.11	0.54	
^{141}Xe	0.02	0.14	0.06	−0.02	
^{141}Cs	0.08	0.17	0.04	0.14	0.06
^{142}I	0.45	0.51	0.47	0.45	
^{142}Xe	0.09	0.29	0.22	0.07	
^{142}Cs	0.14	0.20	0.13	0.18	0.13
^{143}Xe	0.17	0.28	0.23	0.11	
^{143}Cs	0.24	0.35	0.25	0.29	
^{144}Xe	0.19	0.57	0.29	0.22	
^{144}Cs	0.23	0.30	0.25	0.25	

TABLE II *(Continued)*

Nuclide	Garvey *et al.* (Ref. 2)	Myers and Swiatecki (Ref. 3)	Seeger (Ref. 4)	Zeldes *et al.* (Ref. 5)	Wapstra and Gove (Ref. 6)
	$(Q_\beta - B_n)/Q_\beta$ from				
^{145}Cs	0.38	0.42	0.40	0.37	
^{145}Ba	−0.24	−0.12	−0.19	−0.33	
^{146}Cs	0.25	0.37	0.33	0.32	
^{146}Ba	−0.52	−0.05	−0.18	−0.36	
^{147}Cs	0.43	0.51	0.49	0.43	
^{147}Ba	−0.20	0.06	0.00	−0.13	

I. Mass number 133

At this mass number one activity with half-life 1.47 ± 0.07 sec has been found. The half-life agrees with the half-life found for ^{133}Sn by γ counting.[15] Also, several mass formulas predict ^{133}Sn to be a delayed-neutron precursor (cf. Table II).

The isobar ^{133}In is expected to have a very strong neutron branch, but its half-life is probably very short.

J. Mass number 134

Two neutron activities have been found. The short-lived component of half-life 1.04 ± 0.02 may not be identical to the activity of half-life 0.85 ± 0.10 sec attributed to a 0$^-$ isomer of antimony by Kerek *et al.*[16] These authors also assigned a 10.3 sec activity, with five times higher yield, to a 7$^-$ isomer of antimony. This is in line with the expectation that the independent fission yield for the high-spin isomer is larger than that of the low-spin isomer.[17] In the present study, however, the neutron counting rate at saturation was seven times higher for the short-lived component than for the long-lived one. Assuming the neutron branches to be equal, this finding rules out the assignment of the 1.04 sec activity to a 0$^-$ isomer of antimony, and the logical choice is then ^{134}Sn. This conclusion is also supported by the similarity of its delayed-neutron spectrum to that of ^{136}Te. Both ^{136}Te and ^{134}Sn decay to neutron-emitting daughters with 83 neutrons, and all known emitters with one neutron in excess of a closed shell exhibit a pronounced fine structure in their neutron spectra.[18]

The half-life of the short-lived component is in

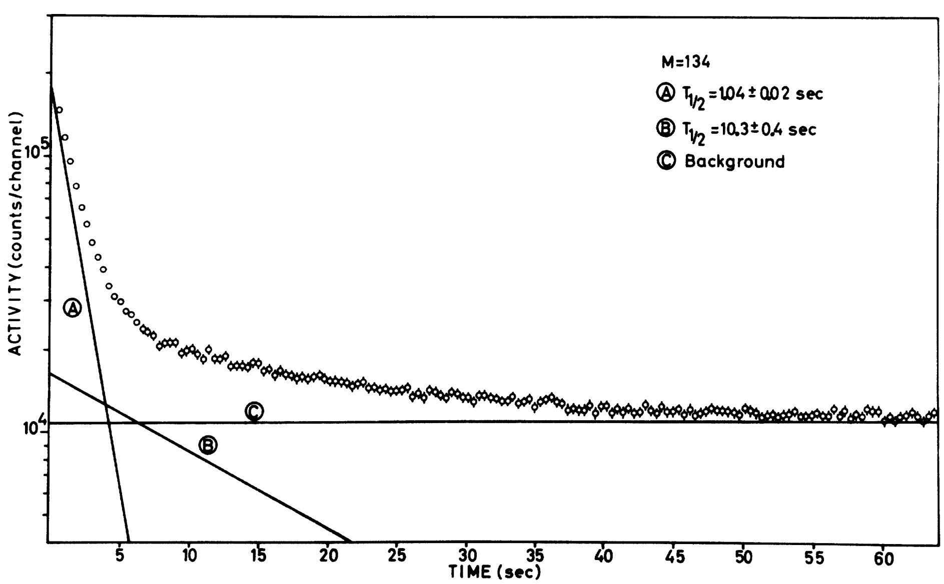

FIG. 1. Decay curve of mass number 134 showing the two components, the short-lived one presumably due to ^{134}Sn and the long-lived one due to ^{134}Sb.

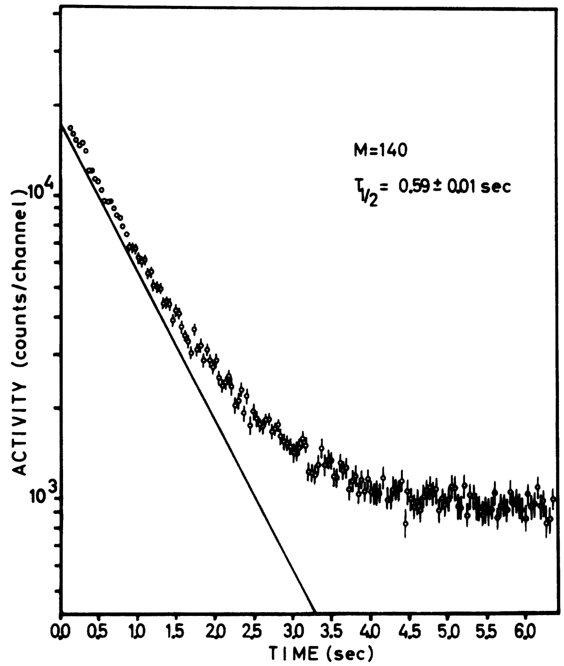

FIG. 2. Decay curve of ^{140}I.

fair agreement with a recent determination by Asghar *et al.*[19] who got the value 0.7 ± 0.2 sec for the half-life of ^{134}Sn. The half-life of the long-lived component agrees well with measurements for the earlier known ^{134}Sb.[16,20,21] A decay curve showing the two activities is given in Fig. 1.

K. Mass number 135

Only one activity has been found, of half-life 1.82 ± 0.04 sec. The discrepancy between this value and 1.70 ± 0.02 sec for the half-life of ^{135}Sb given by Tomlinson and Hurdus[22] may be explained as follows. The latter authors did not use mass separation in their experiment. Consequently, their value might have been affected by contaminations from ^{136}Sb with a half-life of 0.82 ± 0.02 sec (see Sec. II L).

L. Mass number 136

Two neutron activities have been found. An accurate half-life determination yielding 17.5 ± 0.2 sec in this work is in disagreement with the value 20.9 ± 0.5 sec for ^{136}Te obtained from γ counting.[23]

The short-lived activity is found to have a half-life of 0.82 ± 0.02 sec. Antimony and tellurium are both expected to be delayed-neutron precursors. Isomeric states of even-mass Te isotopes are not expected, however, and ^{136}Sb is therefore the most probable assignment of this short-lived activity.

M. Mass numbers 137, 138, and 139

Iodine isotopes of these mass numbers are known to be delayed-neutron precursors. For the half-lives (cf. Table I) there is agreement, within limits of error, with most of the older determinations obtained using neutron counting or β counting. The present result for ^{139}I is more accurate than the older determinations, however.

Traces of the 3.5 sec ^{137}Te precursor[24] were seen in experiments with the collection time chosen so as to enhance short-lived activities, but the activity was too weak to permit a half-life determination.

N. Mass number 140

According to Table II only iodine is likely to be a delayed neutron precursor. We have found one activity with half-life 0.59 ± 0.01 sec. This is attributed to iodine. The half-life is in serious disagreement with the result 0.86 ± 0.04 sec obtained for this nuclide by milking techniques.[25] A decay curve of ^{140}I is shown in Fig. 2.

O. Mass number 141

At this mass number all three elements iodine, xenon, and cesium are reported to be delayed-neutron precursors.[24] We have studied the long-lived component, which should be cesium, and the short-lived one, presumably iodine, but we did not measure the component of intermediate half-life, i.e. xenon. For the long-lived isobar the half-life 22.2 ± 0.2 sec was found. It is lower than the value obtained by β counting[8] or by other delayed-neutron experiments.[26,27] For the short-lived component the half-life 0.48 ± 0.03 sec has been obtained, in agreement with the value 0.43 ± 0.08 sec reported in the literature.[25]

P. Mass number 142

Both ^{142}Xe and ^{142}Cs are known to be delayed-neutron precursors. Their half-lives are similar, 1.24 and 1.89 sec, respectively, and it is difficult to resolve a composite decay curve into its components. However, fission yields and separator efficiencies indicate that there should be about ten times more neutrons from ^{142}Cs than from ^{142}Xe.[28] Thus, the half-life value 1.69 ± 0.09 sec obtained in the present work should be representative for ^{142}Cs. It is in agreement with the earlier value 1.68 ± 0.02 sec[26] obtained by γ and neutron counting.

Q. Mass number 143

Both ^{143}Xe and ^{143}Cs have been reported to be delayed-neutron precursors.[24] Again, the xenon

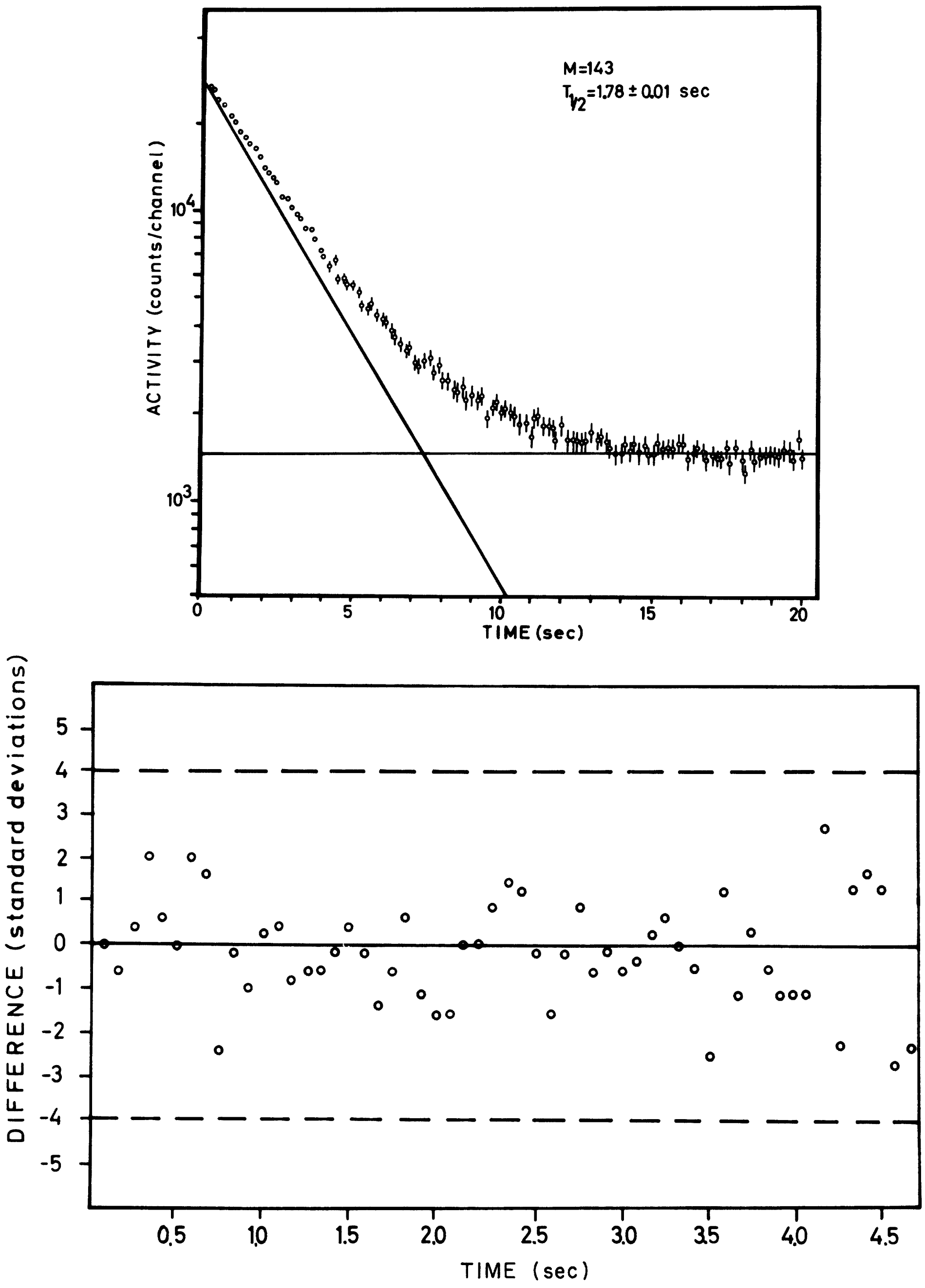

FIG. 3. Decay curve of mass number 143. The lower part indicates the difference between measured and calculated activity under the assumption of only one component.

isotope will probably contribute less than the cesium isotope to the neutron activity.[28] Moreover, the decay curve shown in Fig. 3 does not indicate any short-lived component. The half-life obtained in this work, 1.78 ± 0.01 sec, should thus be representative for ^{143}Cs.

The half-life determination agrees well with the earlier measurements 1.60 ± 0.14,[29] 1.69 ± 0.13,[30] and 1.7 ± 0.1 sec.[31] The accuracy is considerably higher than in these older measurements.

R. Mass number 144

The half-life found at this mass, 1.00 ± 0.02 sec, is in excellent agreement with earlier measurements[29,30] on cesium isotopes. The good statistics in the present experiment has allowed a considerable improvement in the accuracy of the measurement.

S. Mass number 145

The half-life found is 0.584 ± 0.006 sec for this precursor which earlier experiments have shown to be ^{145}Cs.[32,33] It agrees with these measurements, but the accuracy is improved.

T. Mass number 146

One neutron activity is found at this mass number. Its half-life, 0.343 ± 0.007 sec, is in agreement with a recent measurement of the delayed-neutron precursor ^{146}Cs yielding the half-life 0.352 ± 0.042 sec.[33] The discrepancy with an older determination giving the result 0.189 ± 0.011 sec is unexplained.

U. Mass numbers 120, 121, 124, 125, 126, and 147

At mass numbers 120, 124, 125, 126, and 147 none or a very small difference between the beginning and the end of the decay curve has been found. For mass number 121 there is a significant excess of neutrons in the beginning of the multiscale curve which, according to Table II, should be due to ^{121}Ag.

III. SUMMARY

The present study has resulted in the detection of 12 new delayed-neutron precursors: ^{122}Ag, ^{123}Ag, ^{127}In, ^{128}Cd (or $^{128}In^m$), ^{128}In, ^{129}In, $^{129}In^m$, ^{130}In, ^{131}In, ^{132}In, ^{133}Sn, and ^{136}Sb. For ^{134}Sn, ^{135}Sb, ^{136}Te, ^{139}I, ^{140}I, ^{141}I, ^{143}Cs, ^{144}Cs, ^{145}Cs, and ^{146}Cs the accuracy of the half-life determination is improved.

Finally, a strong indication of delayed neutrons from ^{121}Ag has been found.

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