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EXCITATION ENERGY OF THE SPONTANEOUSLY FISSIONING ISOMERIC STATE IN ^{240}Am

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Энергия возбуждения спонтанно-делящегося изомерного состояния Am^{240}

Была измерена функция возбуждения для реакции $Pu^{241}(p,2n)$, приводящей к спонтанно делящемуся изомеру Am^{240} в интервале 9,6–13,6 мэв. Функция возбуждения обнаруживает порог при энергии 10,7±0,1 мэв. Отсюда делается вывод, что энергия возбуждения изомерного состояния равна 3,15±0,3 мэв.

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Excitation Energy of the Spontaneously Fissioning Isomeric State in ^{240}Am.

The excitation function for the reaction $^{241}Pu(p,2n)^{240m}Am$ leading to the spontaneously fissioning isomeric state in ^{240}Am has been measured with proton energies 9.6–13.6 MeV. The excitation function shows a typical threshold behaviour, corresponding to a threshold of 10±0.1 MeV. This is 3.15±0.25 MeV higher than the estimated threshold for the ground state reaction, and it is interpreted as the excitation energy of the isomeric state. Possible reasons for the unusual stability towards γ-emission of such a high lying level are discussed.

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EXCITATION ENERGY OF THE SPONTANEOUSLY FISSIONING ISOMERIC STATE IN ^{240}Am

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1. Introduction

The discovery[1] of isomeric levels of transuranium nuclides, undergoing relatively fast spontaneous fission decay, presents a puzzle in several respects.

In terms of the usual semiempirical concepts of spontaneous fission one would expect the isomeric levels in $^{240,242,244}Am$, with half lives in the millisecond region[2,3], to lie at least 2.7 MeV above their respective ground states[4]. But, on the basis of what is generally known about nuclear isomerism, it is not readily understood how such high lying states can be relatively stable towards γ-emission. Spin values higher than twenty would be required, and this is incompatible with the reaction data[5]. In a recent theoretical investigation Malov et al.[6] show that a detailed consideration of the approximate selection rules and hindrance factors, associated with conservation of the spin projection K and with independent motion of unpaired particles, can explain the possible existence of a long lived excited state in $^{242}_{147}Am^{95}$ with the excitation energy 1.0-1.2 MeV and $K\pi = 12-$. It is also clear from this study that the conditions for stability against on line γ-emission of a state of this type will vanish, if its excitation energy lies just a few hundred keV above this upper limit.

Thus, the spontaneously fissioning isomeric states must have unusual properties, either with respect to their fissionability or with respect to their γ-emission stability - or both.

Clearly, a determination of the excitation energy (and of the spin) of the isomeric level is desirable. For this purpose a measurement of γ-rays or conversion electron transitions to the ground state would be valuable; but most difficult, however, in view of the low partial cross section for the population of the isomer (σ(d,2n; 12 MeV d) $\tilde{\sim}$ 10^{-29} cm^2) and the high background from fission product activities. The possibility of measuring an α-decay branch was considered by Leachman et al.[7], who found, however, a limit of less than one α-particle from ^{242m}Am per hundred spontaneous fissions. Short of adequate direct methods it seems necessary to resort to a more indirect determination. Here, a measurement of the threshold for the reaction

$$^{241}\mathrm{Pu}(p,2n)^{240m}\mathrm{Am};\quad \begin{matrix} Q(\mathrm{gr.st.}) \tilde{\sim} 7.5\ \mathrm{MeV} \\ t_{1/2}(\mathrm{isomer}) \tilde{\sim} 0.6\ \mathrm{ms} \end{matrix} \qquad (1)$$

seems particularly suited, since the expected threshold lies high enough to the Coulomb barrier in plutonium to make measurements near threshold possible.

The present paper describes experiments in which the excitation function for the above reaction is measured. The cross section is found to rise very steeply above 10.6 MeV, and a detailed comparison with the Jackson evaporation model[8] for the (p,2n) reaction, using the appropriate nuclear temperature, defines the reaction threshold to be 10.7±0.1 MeV. A comparison with analogcus excitation functions[19] for the population of excited states in ^{168}Yb where the thresholds are known gives the same result.

The threshold for the ground state reaction (1) is 7.55±0.2 MeV, i.e. the energy difference is 3.15±0.25 MeV. It seems difficult to avoid the conclusion that this is also the excitation energy of the isomeric level.

If so, the stability towards γ-emission must be due to a new effect, and it seems not unreasonable to imagine that the isomer possesses a different equilibrium deformation separated from the ground state by an appreciable energy barrier as previously discussed by Flerov[10]. It is interesting to note in this connection that according to recent theoretical considerations by Strutinsky[11], one may indeed expect the existence of pronounced secondary minima in the nuclear surface of some nuclides, particularly among the heaviest elements.

2. Experimental Procedure

2.1. General Remarks

The measurements have been carried out at the tandem Van de Graaff laboratory of the Niels Bohr Institute in collaboration with the Laboratory of Nuclear Reactions, J.I.N.R. Dubna.

Three series of experiments were performed. The first consisted in a comparison of the reactions

$$^{242}Pu(d,2n)^{242m}Am;\ t_{1/2} = 14\ ms \qquad (2)$$

$$^{240}Pu(d,2n)^{240m}Am;\ t_{1/2} = 0.9\ ms \qquad (3)$$

with 12 MeV deuterons in order to confirm the existence of the isomer in ^{240m}Am and measure its half life accurately.

In the second series, the excitation function and threshold for the main reaction

$$^{241}Pu(p,2n)^{240m}Am \qquad (4)$$

was measured with proton energies 9.6-13.6 MeV. The third series was devoted to measurements of background effects. The ^{241}Pu sample contains 3% ^{240}Pu and 16% ^{241}Am. Therefore the yield of the

$$^{240}Pu(p,n)^{240m}Am \qquad (5)$$

and the

$$^{241}Am(p,d \text{ or } p,pn)^{240m}Am \qquad (6)$$

reactions were measured, using highly enriched targets of ^{240}Pu and ^{241}Am.

Fig.1 shows the irradiation chamber schematically. The target is activated with proton pulses of 1.9 ms duration and detectors of ordinary glass plate[12,13] are used to record delayed fission fragments in the 3.2 ms periods between the beam pulses. The beam is pulsed by electrostatic deflection[14]. Synchronisation between the motion of the wheel and the beam is achieved with a photocell arrangement and slits in the wheel, fig.1. Electromagnetically separated plutonium isotopes were obtained from A.E.R.E., Harwell.

2.2. Target Preparation

All targets were prepared by the molecular plating methods[15,16]. The plating solutions were made up of isopropanol containing plutonium as the nitrate dissolved in a minimum amount of dilute nitric acid. Plating voltages were

400-600 V, giving current of 0.5-0.8 mA across the circular target area of 0.12 cm^2. The target material was plated onto 200 $\mu g/cm^2$ Ni foils with a supporting Cu-layer on the back side. The Cu-layer was removed by dissolution in ammoniacal trichloroacetic acid after mounting the foil on the 10×12 mm^2 target frame.

The exact chemical form of the deposit is uncertain, most probably it is a mixture of plutonium oxide and nitrate. Typically, it is brownish of colour, has a rather smooth surface and adheres well to the backing material.

The isotopic composition of the target material, based on the specification of the supplier, A.E.R.E., Harwell, and corrected for decay of ^{241}Pu into ^{241}Am, is shown in table 1.

Independently, a mass analysis of aliquots of the various Pu samples was performed at Eurochemic, Belgium. Prior to the analysis the Pu was isolated from Am (and possible U) impurities. The results are given in parenthesis in table 1 and show good agreement.

No chemical fractionation between Pu and Am is assumed to take place in the target preparation process.

Target thicknesses were determined by α-counting using a surface-barrier silicon detector with known efficiency. The solid angle subtended by the detector was known both from a measurement of linear dimensions and from comparison with absolute count rates of representative samples obtained with a windowless gass-flow 2π proportional counter. Knowing, furthermore, the target

area and the isotopic composition the average thickness of the heavy element constituent of the targets can be calculated. They were typically 100-400 $\mu g/cm^2$. The combined effect of uncertainties in α-counting, detector geometry, and target area together with the influence of a possible uneven distribution of the target deposit is estimated to result in an over-all uncertainty of 20-25% in the determination of the true target thickness as felt by the beam.

The glass detectors were developed after exposure together with a test plate previously exposed to a weak. ^{252}Cf source, by etching with 2.5% hydrofluoric acid solution for about 30 minutes at room temperature. This time was chosen to obtain a diameter of 7-8 microns for tracks that were perpendicular to the surface. The plates were subsequently washed under running water for 20-30 minutes and dried.

The area of the plates which was exposed to fission fragments was marked with the aid of a glass diamond and likewise divided into four equal zones. The number of tracks in each zone was counted under microscope with 10×12.5 magnification. All exposures were counted twice; once at the Niels Bohr Institute and once at the Joint Institute

2.3. Evaluation of Cross Sections

Delayed fission: The efficiency of detecting fission fragments with the glass plates depends on the following factors: solid angle, half life of the isomer, rotation frequency of the wheel, angular size of the glass plates and

the efficiency of the developing and scanning procedure.

To determine the solid angle the wheel with glass plates was run for a definite time with the target replaced by a ^{252}Cf source of similar shape and size. The spontaneous fission count rate of the source had previously been determined using a windowless gas-flow 2π proportional counter having a reasonably good plateau for fission fragments. The solid angle was found to be $(0.365 \pm 0.03)/2\pi$.

The efficiency factor correcting for loss due to decay of the isomer is determined graphically. For the two half lives in question, the result is 0.345 (0.91 ms) and 0.423 (14 ms). (The wheel moves through 90° in 5.05 ms.)

To be able to calculate cross sections one also has to know target thicknesses (see above) and the integrated beam on the target, which is measured as the charge collected by the Faraday cup.

The over-all uncertainty in the determination of absolute isomer cross sections is estimated to be about 30%.

Prompt fission: The prompt fission fragments were recorded during the beam periods by a surface-barrier silicon detector mounted at an angle of about 41° with respect to the beam direction, fig.1. The detector geometry was determined both by a direct geometrical measurement and by α-counting of a sample with known count rate. The solid angle was found to be $\Omega = (2.47 \pm 10) \times 10^{-4}/4\pi$. For detection of fission events the effective geometry is twice this value.

Knowing the target thickness and the integrated beam

an average prompt fission cross section can be calculated for each target. The results are given in table 2.

In calculating the prompt fission cross sections the fission fragment angular distribution is assumed to be isotropic, although this is not strictly so. A comparison with angular anisotropy data for similar reactions[14,13] indicates, however, that the error introduced in this way is insignificant, compared to the over-all experimental uncertainty of about 30%. It should be noted that the detector angle corresponds to a position near the midpoint between the extrema of the angular distribution.

2.4. Decay Curves

As stated the glass plates were scanned in four equal zones corresponding to consecutive time periods. A four-point decay curve can thus be drawn for each exposure to check that the fission really is due to isomer decay and not to ground-state spontaneous fission or to stray neutrons causing prompt fission in the beam-off periods. All exposures show a clean decay, except the four lowest-energy irradiations of ^{241}Pu (see table 2) where a constant background of the order of 20% is present. Presumably it is due to the $^{242}Pu(p,n)^{242m}Am$ reaction with the 0.50% ^{242}Pu contamination present in the target.

3. Results

3.1. The half life of ^{240m}Am

In fig.2 the results of the decay measurements are shown for the reactions (3), (4) and (5) separately. It is seen that the half lives are identical within the limits of the statistical error. The weighted mean value is

$$t_{1/2}(^{240m}\mathrm{Am}) = 0.91 \pm 0.07 \text{ ms}$$

in fair agreement with the result of Polikanov et al.[3], 0.60±0.2 ms.

3.2. Cross sections

The measured cross sections for prompt and delayed fission are shown in table 2. The largest contribution to the experimental uncertainty comes from the uncertainty in estimating target thickness.

Within the estimated experimental uncertainty of 30% the prompt fission cross sections are consistent with the measurements of Huizenga and Vandenbosch[17] and others[18] for nearby target nuclei.

The delayed fission cross section for the reaction (2) agrees with the result of Brenner et al.[15], $\sigma(d,2n) = 8 \pm 3$ μb for 12 MeV d.

In fig.3 the result of proton bombardment of the ^{241}Pu targets are shown. In addition to the measured prompt and delayed fission cross sections, the measured background contribution from the ^{240}Pu(p,n) reaction, normalized to the 3.1% abundance of this isotope in the ^{241}Pu target, is shown. The background contribution from the reaction (6) is zero, table 1. Also shown in fig.3 is the theoretical

curve for the total compound nucleus cross section, σ_T, from an optical model calculation due to Jägere[19].

3.3. Threshold of the $^{241}Pu(p,2n)^{240m}Am$ Reaction

The dramatic decrease in the cross section for production of the ^{240}Am isomer at proton energies 11.6-10.6 MeV is evidently a threshold effect. The available evidence indicates that the fission isomer is formed by a compound nuclear reaction. We will therefore analyze the results in terms of the Jackson neutron evaporation model[8].

The cross section of a (p,2n) reaction (or any other compound nucleus reaction with the boil-off of two neutrons) in the interval between its threshold and the next higher threshold (for (p,3n) or (p,2nf) reactions) can be expressed [20,8] as

$$\sigma(p,2n) = \sigma_T \times p \times [1-(1+\Delta E/T)\exp(-\Delta E/T)] \qquad (7)$$

where p is a branching ratio, taking into account competition between fission and neutron evaporation as well as neutron evaporation processes, which energetically might lead to the isomer but actually bypasses it; ΔE is the energy above threshold $E_p - E_{th}$; and T is the nuclear temperature. It is considered to be a constant in accordance with the usual assumption of the Jackson model.

In the relatively small energy interval 10.6-13.6MeV remote from other thresholds, p is assumed to be constant. Rewriting (7) therefore gives

$$\sigma(p,2n)/\sigma_T \sim 1-(1+\Delta E/T)\exp(-\Delta E/T) \qquad (8)$$

In fig.4a the left hand side of (8), determined as described in subsection 3.2, is compared with a calculated curve corresponding to the right hand side of (8), using a nuclear temperature T = 1.35 MeV, found to be appropriate to experimental excitation functions for ^{235}U + α-particles[21] and the threshold energy E_{th} = 10.70 MeV. The curve is normalized to the experimental value at 11.6 MeV bombarding energy. If, instead of T = 1.35 MeV, values of T = 1.0 MeV or T = 2.0 MeV are used, the fits to the experimental points are slightly less satisfactory, but the value of E_{th} required to give a best fit only changes by 0.1 MeV. Within the assumptions made here, one can therefore safely say that the experimental excitation function determines the threshold to be

$$E_{th}(\text{isomer}) = 10.70 \pm 0.1 \text{ MeV}.$$

In order to test the assumptions for the threshold behaviour we have compared with reaction data for the reaction

$$^{169}Tm(p,2n)^{168}Yb \qquad (9)$$

which has been investigated by R.Graetzer et al.[9]. In this case the threshold for the ground state reaction is known[22]. The authors have measured excitation functions for excited states, whose energy above the ground state they determined through measurements of the deexciting conversion electrons. In this case $T = 1.35(240/168)^{1/2} = 1.61$ MeV is used. The comparison is shown on fig.4b. The fit is satisfactory and apparently insensitive to the effect of spin and excitation energy, i.e. level density.

It should be noted that the conclusion about the threshold value does not in any essential way depend on the application of eq.(8). The threshold value which one derives will not change significantly if a simpler phenomenological comparison of the excitation functions for the reactions (1) and (9) is used as the basis for the evaluation. Thus, there seems, altogether, to be good reasons to believe that present experiment determines the threshold energy with the accuracy ±0.1 MeV.

4. Discussion

The Q-value for the $^{241}Pu(p,2n)^{240}Am$ ground state reaction is not accurately known experimentally; the reason being that there only exists an estimated value for the electron capture decay energy of ^{240}Am. This estimate[23], 1300 keV, is equivalent to a neutron binding energy of ^{241}Am, B(N,241) = 6.7 MeV. By extrapolation from ^{243}Am and ^{245}Am, where the neutron binding energies are accurately known[22], a value of B(N,241) = 6.6 MeV is predicted. In this region of isotopes the neutron binding energies change 0.2-0.3 MeV when going from nucleus A to (A-2) with the same Z-value. The extrapolation may therefore be assumed to be accurate within ±0.2 MeV and hence, the mass of ^{240}Am is determined with the same accuracy. Applying a 0.05 MeV correction for recoil energy one obtains[22]

$$E_{th}(gr.st) = 7.55 \pm 0.2 \text{ MeV}.$$

The difference between thresholds

$$E(exct) = 3.15 \pm 0.25 \text{ MeV}$$

then represents the upper limit for the excitation energy of the spontaneously fissioning isomer in ^{240}Am. The actual energy may in principle be smaller, because one could imagine the isomer to be accessible only by way of γ-emission following the neutron evaporation. Considering, however, (I) that the compound nucleus reaction excites all the degrees of freedom of the nucleus, (II) that the level density at 3 MeV excitation energy is quite high in an odd nucleus, and (III) that the cross section is found to decrease three orders of magnitude without any indications of sub-threshold effects, it seems very unlikely that the isomer could have an excitation energy lower than 3.15 ± 0.25 MeV.

Since very high spin values ($I > 20$), required to explain the γ-emission stability of such a high lying isomeric level, are excluded by the reaction data[5], it follows that one cannot apply usual spin distribution estimates[24] to explain the experimental isomer ratio. The usual treatment is based on the assumptions that (I) the energy difference between the ground state and the isomer level is small and does not influence the isomer ratio, and (II) the isomer cross section represents the cumulative yield of all reactions going through high-spin channels above a certain limiting spin value (or inversely if the spin of the isomer is lower than the ground state spin). In the present case, none of these conditions apply. Thus, it is trivial that the isomer ratio must go to zero when the threshold is

approached; but it is also to be expected that the tendency to accumulate reaction yield on the isomeric state through γ-radiation from higher lying states is seriously affected by the existence of lower lying levels, which compete for the γ-rays. Analogous considerations apply to the population frequency by direct neutron evaporation. Recently Sommer and Prokofjev[25] have considered the problem of calculating isomer ratios as a function of excitation energy of the isomeric level by a statistical approach which disregards all possible selection rules, including spin selection rules, and only takes into account the energy dependence of neutron evaporation and γ-emission probabilities. Applying such formalism to ^{242}Am, using realistic data for the level density, they conclude that the experimentally measured isomer ratio, $\sigma(\text{isomer})/\sigma(\text{gr.st}) = 4\times10^{-4}$ for the reaction (2) with 10-13 MeV deuterons, corresponds to an excitation energy of 2.8 MeV.

This result indicates that the experimental reaction cross sections are quite compatible with an excitation energy for the isomer of theorder of 3 MeV. Such a conclusion requires, however, that the measured cross section are representative, viz., that spontaneous fission is the predominant decay mode. Experimental proof hereof is still lacking. (Thus F.S.Stephens et al.[26] find only a limit $\sigma(\text{s.f.})/\sigma(\gamma) > 10^{-2}$, whereas $\sigma(\text{s.f.})/\sigma(\alpha\text{-decay}) > 10^{2}$, ref.[7].)

The enhanced probability for spontaneous fission is readily understood if the excitation energy is 3 MeV. It

remains a puzzle why γ-emission is so highly retarded ($t_{1/2}(\gamma) > 0.91$ ms).

When it is considered that the nuclear energy surface for the heaviest nuclei is relatively shallow, it may not be unreasonable to imagine that there can exist secondary minima in the energy surface, separated from the ground state minimum by an appreciable energy barrier, and that the fissioning isomers are nuclei caught in such a minimum.

Swiatecki et al. have shown[27] that the experimentally observed deviations from the liquid drop predictions of nuclear binding energies might be understood as the result of inhomogenuities in the single-particle level densities, associated with shell structure; since the liquid drop formula is based on the assumption of a homogenuous level density distribution in the nucleus. A high level density at the Fermi surface results in a smaller binding energy and, hence, some nuclei may be more strongly bound in a nonspherical equilibrium configuration, where the level density is nearer to the average liquid drop level density; despite the increase in surface energy. Strutinski, carrying this kind of approach further, has analysed level densities in the Nilsson scheme[28] as a function of mass number and of deformation. He finds[11] that although there is a general tendency to approach a constant level density with increasing deformation, significant fluctuations may persist at large deformations. As a result secondary minima will occur at large deformations in some nuclei.

The existence of spontaneously fissioning isomers may very well be a reflection of such effects. There seems to be hope of a more quantitative understanding of the phenomenon.

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Table 1. Isotopic composition of the targets.

Target	Percent of isotope			
	^{239}Pu	^{240}Pu	^{241}Pu + ^{241}Am	^{242}Pu
^{240}Pu	1.91	98.0	0.074	0.019
	(0.95)	(98.9)	(0.060)	(0.051)
^{241}Pu	1.5	3.09	78.8 + 16.1	0.49
	(1.6)	(3.14)	(78.8 x))	(0.50)
^{242}Pu	3.13	2.90	3.09	90.88
	(3.03)	(2.29)	(2.54)	(92.13)

x) Normalized to 78.8%.

Table 2 For the $^{241}Pu(p,2n)^{240m}Am$ reaction only relative errors are given. The total uncertainty in all the cross section determinations is estimated to be 30%. The total number of delayed fission tracks, column 3, is listed to give an idea of the counting statistics. It is not directly proportional to the cross section because it refers to targets of varying thickness and to different duration of the irradiations.

Reaction	Bombarding energy	Total no.of delayed fission tracks	Cross sections	
			Prompt fission	Delayed fission
	(MeV)		($10^{-27}cm^2$)	($10^{-30}cm^2$)
$^{242}Pu(d,2n)^{242m}Am$	12.1	15 558	170	7.6 ±2.0
$^{240}Pu(d,2n)^{240m}Am$	12.1	5 918	215	3.6 ±1.5
$^{241}Pu(p,2n)^{240m}Am$	9.6	114	29	0.012±0.007 x)
	10.1	57	50	0.005±0.007 x)
	10.3	157	58	0.006±0.007 x)
	10.6	98	73	0.010±0.007 x)
	10.9	257	90	0.13 ±0.02
	11.1	196	105	0.52 ±0.08
	11.3	810	116	0.88 ±0.18
	11.6	1 602	139	1.9 ±0.4
	12.1	3 377	178	4.0 ±0.4
	12.6	4 282	234	10.5 ±2.0
	13.6	744	350	19.4 ±2.4
$^{240}Pu(p,n)^{240m}Am$	10.3	464	-	0.37 ±0.11
	10.9	355	94	0.44 ±0.13
	11.3	174	105	0.28 ±0.09
$^{241}Am\{(p,d)\ (p,p'n)\}^{240m}Am$	10.3	0	-	<0.004 xx)

x) Corrected for a 20% flat background observed in the decay curves, and for the contribution from the $^{240}Pu(p,n)^{240m}Am$ reaction, due to the ^{240}Pu contents of the ^{241}Pu targets.

xx) Assuming a maximum of 3 delayed fission tracks.

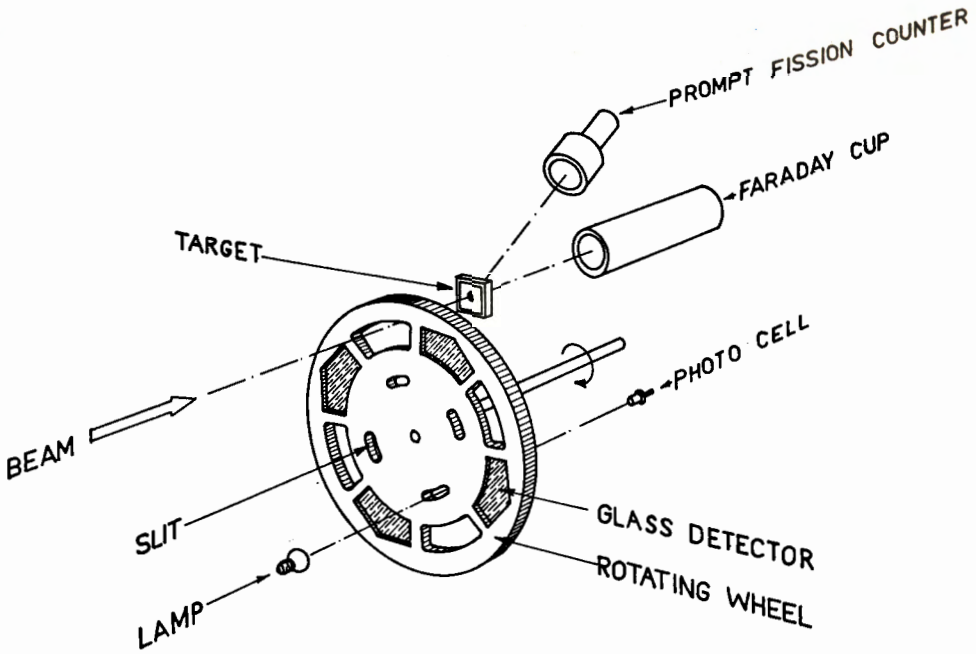

Fig.1 Schematic diagram of the irradiation chamber. The target is stationary. The rotating wheel serves three purposes (I) it brings the glass detectors recording delayed fission fragments in front of the target in the intervals between the beam bursts, (II) by the aid of the slits and the photocell, it provides a pulse which is used to switch the beam off just before the glass plate moves in front of the target, and (III) it prevents any residual beam which may persist in the beam-off periods from hitting the target. The distance between the target and the wheel is 0.5 cm. A collimator (not shown) prevents prompt fission fragments from reaching the glass plates. The target backing is thick enogh to stop recoils, but allows fission fragments to pass through. The beam is monitored by the aid of the Faraday cup as well as with the prompt-fission semiconductor counter.

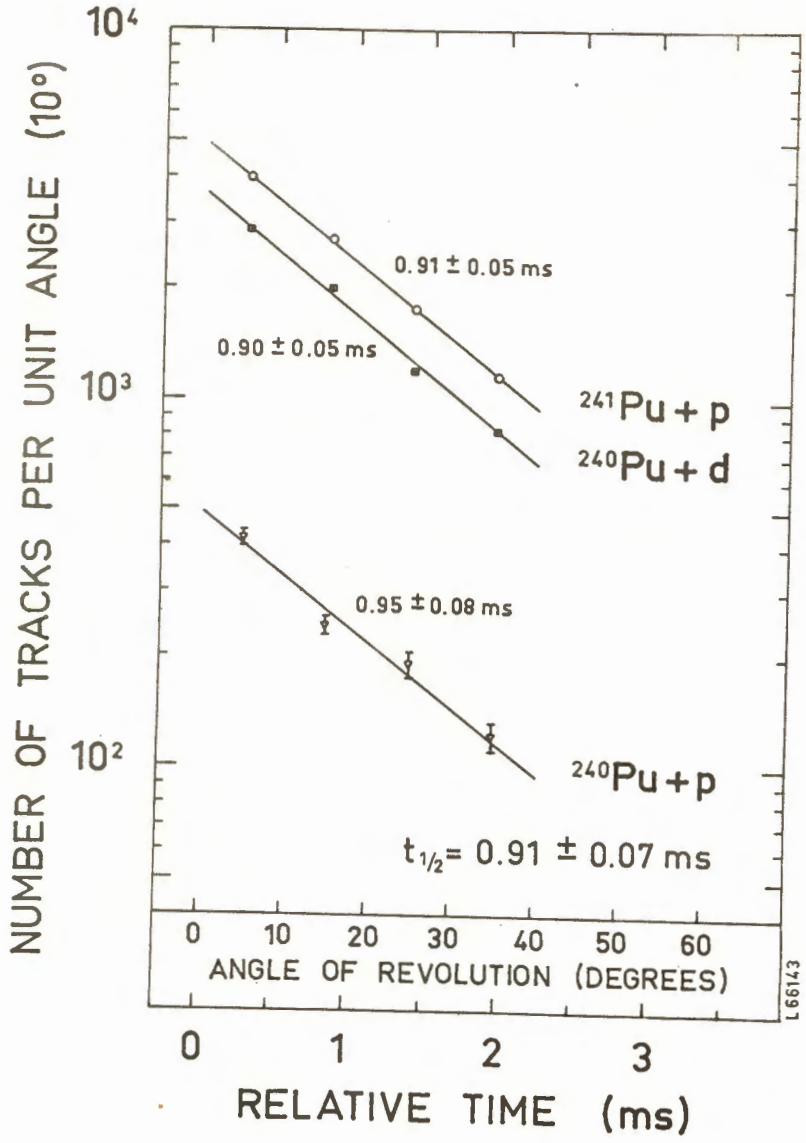

Fig.2 Decay curves for ^{240m}Am produced by three different reactions. The conversion from angle of revolution to time is based on measurement of the rotation frequency of the wheel.

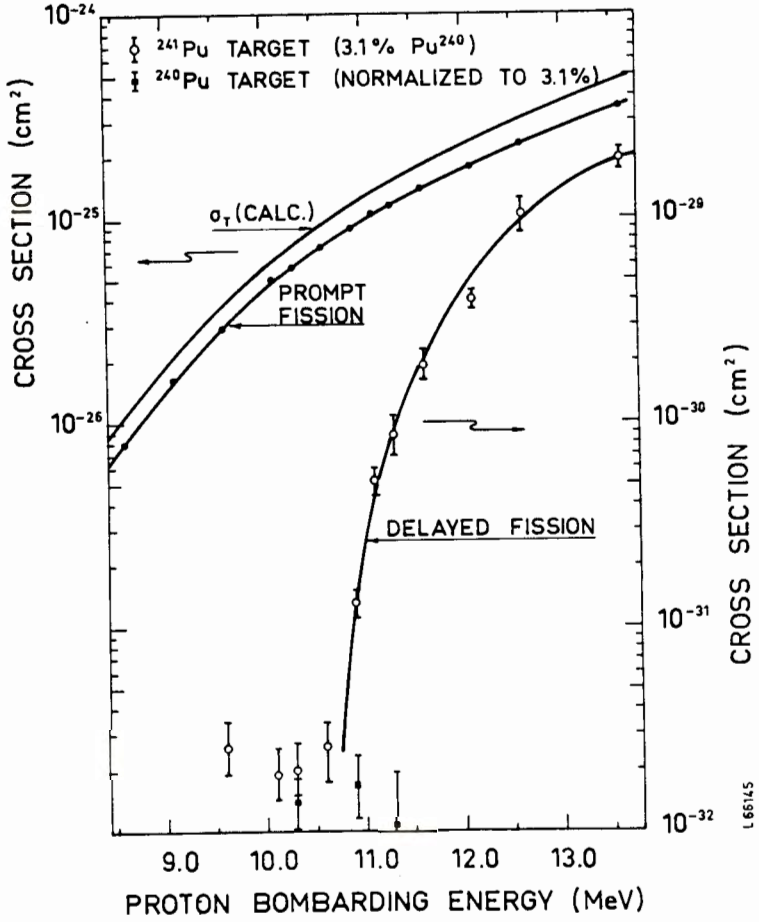

Fig.3 Excitation function for prompt and delayed fission resulting from proton bombardment of ^{241}Pu, see text.

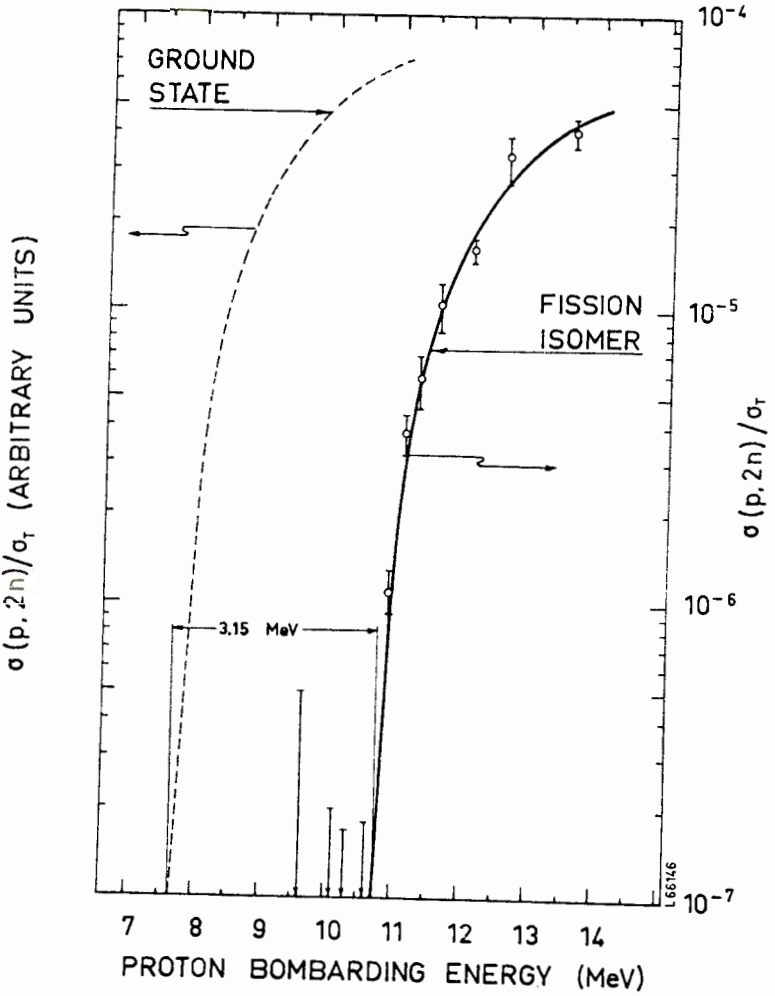

Fig.4a Plot of $\sigma(\text{isomer})/\sigma_T$ for the reaction $^{241}Pu(p,2n)^{240m}Am$ after correction for background. The full drawn curve is calculated from eq.(8), see text. The dotted curve illustrates σ_0/σ_T for the ^{240}Am ground state.

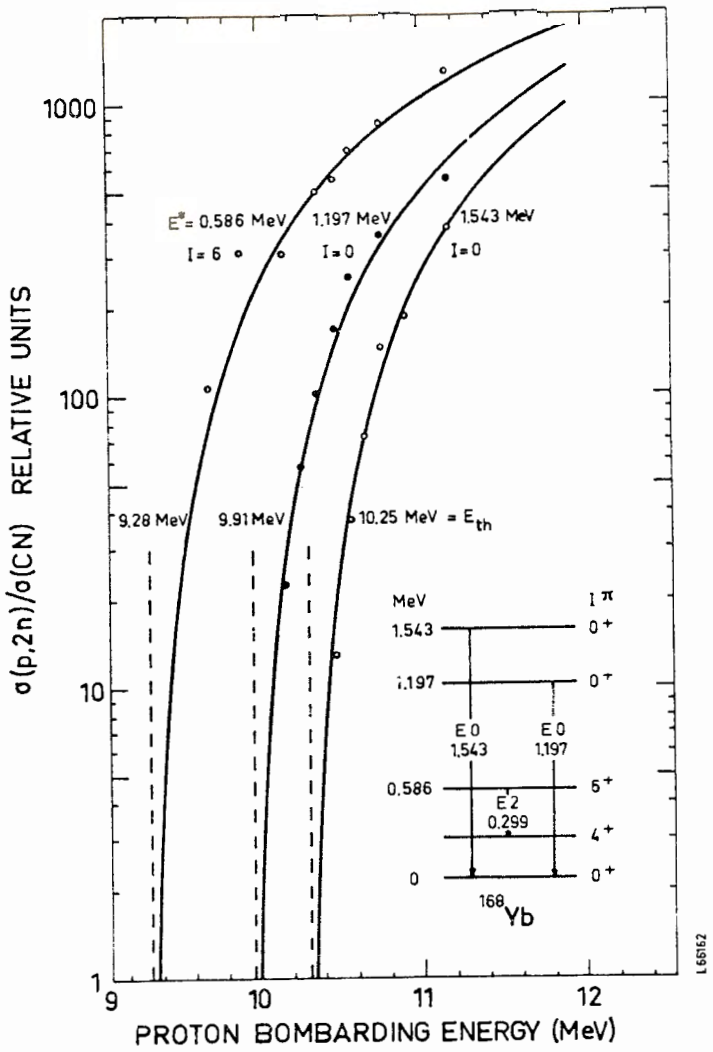

Fig.4b Plot of $\sigma(\text{exct.st.})/\sigma_T$ for the reaction $^{169}Tm(p,2n)^{168}Yb$ leading to various excited states, cf. insert. The threshold energies are corrected for recoil effect.