SEARCH FOR SPONTANEOUSLY FISSIONING ISOMERS PRODUCED WITH 600 MeV PROTONS

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Abstract

Experiments have been performed to produce spontaneously fissioning isomers in the interaction of 600 MeV protons with elements like uranium, thorium and bismuth. No such isomers could be detected having an assumed half-live of 10 sec $\leq T_{1/2} \leq$ 20 d, the upper limit for the production cross-section is between $10^{-32} \geq \sigma \geq 10^{-34}$ cm^2. Furthermore, no spontaneously fissioning isomers could be observed for nanosec half-lives ($\sigma \leq 10^{-30}$ cm^2).

I. Introduction

The phenomenon of spontaneous fission from an isomeric state in isotopes of the elements U, Pu, Am, and Cm has been of considerable interest during the past few years, both experimentally and theoretically. In order to establish the region of half-lives and nuclides where spontaneously fissioning isomers (called "isomers" in this work) can be observed experimentally, various elements have been bombarded with a great variety of particle beams.

With this "search-for" approach it was decided in 1967 to use 600 MeV protons as bombarding particles to determine if isomers could be produced. Particular emphasis was placed on the investigation of long-lived isomers (10 sec $\leq T_{1/2} \leq$ 20 days) produced in uranium and to some extent in bismuth and thorium. High-energy protons are well suited for such a "search-for" program, as quite a large number of spallation products are formed in reasonable yields. (Note however, that in case isomers had been observed, the identification of its nuclear charge and mass would have been difficult.)

The negative results of the experiments reported in Section 2 of this paper can be well understood in terms of the "double-humped" fission barrier model. A general review of this model and the related experiments is given in Ref.1.

Finally, an experiment is described in Section 3 to search for nano-sec isomers. At first, the technique of Ruddy and Alexander[2]), giving apparently incorrect results, was used. Later a slightly modified experimental technique was employed and no isomers could be identified - a result similar to recent findings of Alexander[3]).

II. Search for isomers with 10 sec $\leq T_{1/2} \leq$ 20 d.

1.) Experiments for $T_{1/2} \simeq$ 10 - 20 d and discussion.

Targets of $^{235}U_3O_8$, $^{232}ThF_4$ and ^{209}Bi were prepared on Al-foils and irradiated together with proper Al-monitor foils in the extracted 600 MeV proton beam of the CERN-SC. The irradiations were carried out parasitically with ISOLDE experiments and lasted between 24 h and 96 h. Afterwards, the targets were transported to Marburg and counted for a long time. Fission fragments were counted either with a semiconductor detector in conjunction with a multichannel analyser or with a methane-flow proportional counter operating at 450 V. Some further experimental details are given in Table 1. The loss of fission fragments due to absorption in the "thick" targets was estimated according to Ref.4 and amounted to 20% for a 2.7 mg U_3O_8/cm^2 target. The counter was shielded against neutrons with cadmium and paraffin in the ^{235}U-experiment.

As can be seen from Table 1, the total number of events observed was so small that only upper limits for the production cross-section σ of an isomer with a conveniantly chosen half-live could be given. When the total number of observed events was N = 0 or 1 the calculation was carried out using N = 2. For $2 \leq N \leq 9$, the background was typically very low and the observed number of events was used to estimate σ . In the case of $N \geq 10$, the observed background was substracted. The proton flux was determined by the measurement of ^{22}Na

Table 1: Some experimental details

Target	assumed T 1/2	length of irradiations	start counting after	counting time	repetitions	observed number of events	target thickness (atoms/cm^2)	proton flux per sec	upper limit σ (cm^2)
$^{(nat)}$U	10 sec	60 sec	4 sec	60 sec	66	1	$6.4 \cdot 10^{18}$	$3.8 \cdot 10^{10}$	$3.5 \cdot 10^{-32}$
	1 min	1 min	4 sec	4 min	100	10 2)	$6.4 \cdot 10^{18}$	$3.8 \cdot 10^{10}$	$1.7 \cdot 10^{-32}$
	1 min	1 min	4 sec	2 min	100	12 3)	$5.4 \cdot 10^{18}$	$1.6 \cdot 10^{10}$	$5.7 \cdot 10^{-32}$
	20 min	140-190 min	5 min	40 min	4	0	$6.4 \cdot 10^{18}$	$1.2 \cdot 10^{11}$	$1.6 \cdot 10^{-33}$
	20 min	25.8 h	7 min	200 min	1	2	$6.4 \cdot 10^{18}$	$5.9 \cdot 10^{10}$	$1.1 \cdot 10^{-32}$
	10 h	25.8 h	7 min	27 h	1	14	$6.4 \cdot 10^{18}$	$5.9 \cdot 10^{10}$	$2.9 \cdot 10^{-33}$
	10 h	16 h	5.5 h	16 h	1	6	$6.4 \cdot 10^{18}$	$4.0 \cdot 10^{10}$	$5.5 \cdot 10^{-33}$
	10 h	10.5 h	3 h	10 h	1	0	$6.4 \cdot 10^{18}$	$8.2 \cdot 10^{10}$	$8 \cdot 10^{-34}$
^{235}U(90%)	10 d	100 h 1)	0.5 d	16.8 d	1	6	$1.8 \cdot 10^{19}$	$5.9 \cdot 10^{10}$	$1.4 \cdot 10^{-34}$
	10 d	92 h 1)	8 d	13 d	1	1	$1.8 \cdot 10^{19}$	$9.8 \cdot 10^{10}$	$5.2 \cdot 10^{-35}$
^{232}Th	20 d	24 h	14 d	40 d	1	6	$6.8 \cdot 10^{17}$	$2.5 \cdot 10^{11}$	$1.9 \cdot 10^{-33}$
^{209}Bi	20 d	24 h	2d	58 d	1	27 4)	$9.2 \cdot 10^{17}$	$2.5 \cdot 10^{11}$	$9 \cdot 10^{-34}$

1) the irradiation time was distributed over a period of approximately 10 days.

2) the measured background of 5 events is substracted for the calculation of σ.

3) the measured background of 6 events is substracted for the calculation of σ.

4) the measured background of 12 events is substacted for the calculation of σ.

produced in Al-monitor foils. The experimental results are given in Table 1 and summarized in Table 2 and Fig.1.

The negative results are not surprising in the light of our present understanding of isomer fission (Ref.1). However, the results for Bi might have some importance in the present search for superheavy elements in Nature. Flerov and Perelygin[5]) discuss the possibility, that the apparently very small spontaneous fission activity in some Pb-samples might be due to either the existence of eca-Pb in Nature or to "isomer formation after beta-decay of nuclei". This unlikely type of isomer formation has not been observed so far, including the results of this work.

2.) Experiments with 10 h $\geq T_{1/2} \geq$ 10 sec and discussion.

These experiments were carried out with a natural U_3O_8-target in the beam tube transporting 600 MeV protons from the CERN-SC to the ISOLDE experimental area. As can be seen in Fig.2, a thin target was placed in the proton beam and the irradiations were performed parasitically with ISOLDE runs. After a proper irradiation time the beam was turned off. The target was then lowered and placed in front of a semiconductor fission fragment detector. Using standard electronic equipment the fission fragment counting rate was determined. The geometrical counting efficiency of approx. 50% was determined with a calibrated ^{252}Cf-source.

The results are included in Table 1. In this "beam-off" operation mode the assumed half-live range for $T_{1/2}$ = 20 min and 10 h was investigated, again with negative results.

The study of shorter half-lives (10 sec $\leq T_{1/2} \leq$ 1 min) was carried out with the cyclotron beam operating in the "fast extraction" mode. The transportation of the target from the irradiation position to the counting detector took 4 sec and was carried out by remote control. Since the background was too high during the proton extraction period, the counting system was turned off for 1,8 msec (proton extraction time) every 18 msec. This was accomplished with standard gating

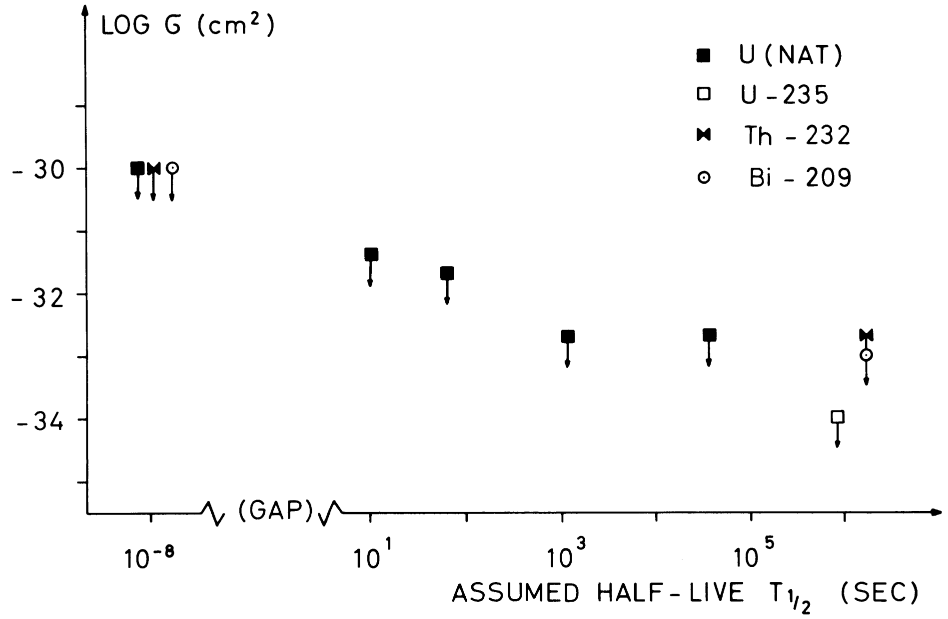


Fig. 1 Upper limits for the production cross-section σ.

Table 2: Summary of the experimental results: Lower limits for the production cross-section of isomers.

Target	assumed half-live	number of experiments	σ (cm^2)
$^{(nat)}$U	10 nsec [1]	2	10^{-30}
^{232}Th	10 nsec [1]	2	10^{-30}
^{209}Bi	10 nsec [1]	2	10^{-30}
$^{(nat)}$U	10 sec	66	$4 \cdot 10^{-32}$
	1 min	200	$2 \cdot 10^{-32}$
	20 min	5	$2 \cdot 10^{-33}$
	10 h	3	$2 \cdot 10^{-33}$
^{235}U	10 d	2	$1 \cdot 10^{-34}$
^{232}Th	20 d	1	$2 \cdot 10^{-33}$
^{209}Bi	20 d	1	$1 \cdot 10^{-33}$

[1] the estimation of the half-live is described in the text

techniques as shown in Fig.3. Additionally, the detector was heavily shielded against neutrons with Cd, paraffin, water and concrete blocks as shown in Fig.2. With this electronic gating and physical shielding, the background with the target in front of the detector was app.1 event/100 min at a beam intensity $(1-4)\cdot 10^{11}$ p/sec. Typically, an experimental cycle consisted of a 1-min irradiation period and several minutes counting. Such a cycle was repeated many times. Again, not even a slight indication of a spontaneous fission activity was detected. The results are included in Fig.1 and Table 1 and 2.

A careful search was made for the 60 sec spontaneous fission activity of ^{227,228}Np produced in the reaction (^{209}Bi + ^{22}Ne) as reported by Kuznetsov, et al.$^{6)}$. However, their production cross-section of $4\cdot 10^{-34}$cm^2 at the peak of the exitation function with ^{22}Ne was considerable lower than our detection limit ($\sigma \leq 10^{-32}$).

The discussion of the negative results has already been given at the end of Section II.,1.).

III. Search for "nanosec" isomers$^{7)}$.

It is known that there are several isomers among uranium isotopes having half-lives in the nanosec range$^{1)}$. Furthermore, it was thought at the time being that isomerism could be observed in many heavy nuclides lighter than uranium. Therefore, it was of interest to see, whether isomers could be produced in detectable quantities by 600 MeV protons on U, Th and Bi.

In the first series of experiments the technique of Ruddy and Alexander$^{2)}$ was used with slight modifications. Fig.4 shows the experimental setup. A closed chamber was evacuated and placed in the short open air section of the proton beam before it reaches the ISOLDE target area$^{8)}$. The targets consisted of natural UF_4, ThF_4 and Bi evaporated onto Al-foils. Pre-etched mica detectors were placed 3 mm downstream from

Fig. 2 Experimental Setup for Isomer Search

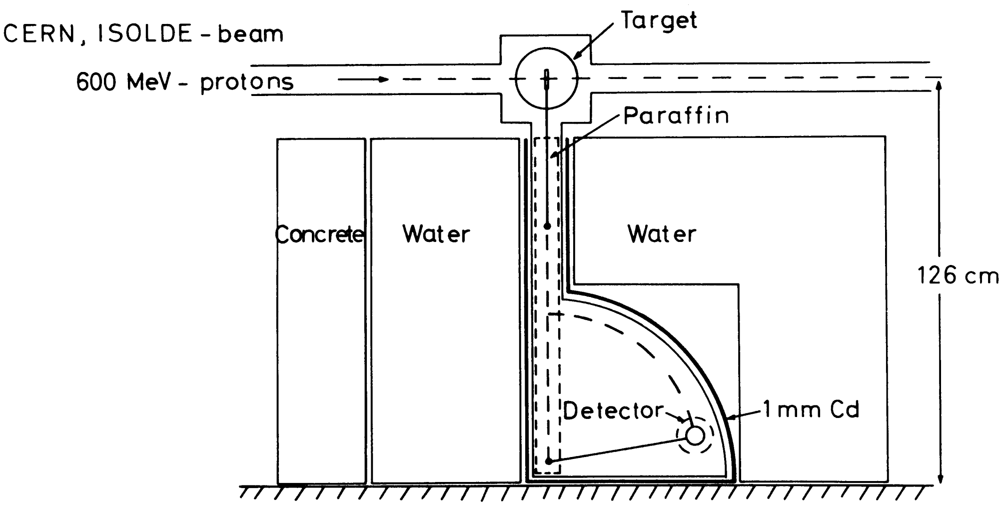


Fig. 3a Pulse Frequency of the Beam at Fast Extraction

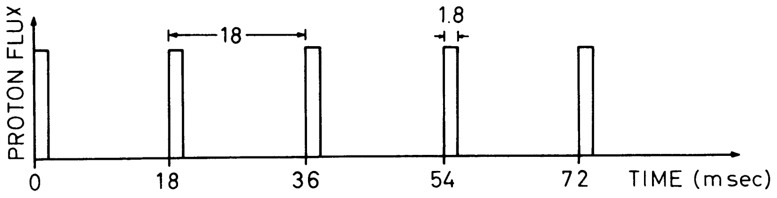
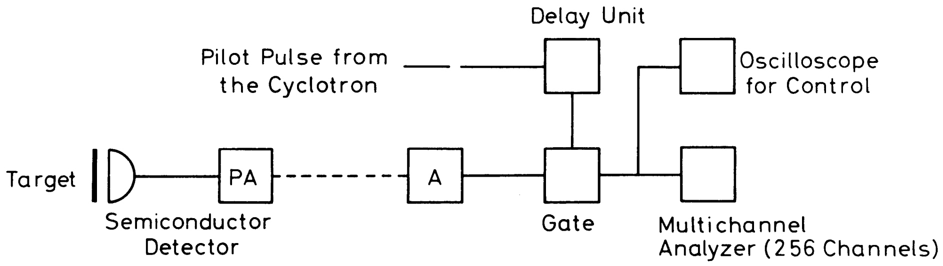


Fig. 3b Block Diagram of the Measuring Setup



the target. In this way the detectors were shielded from prompt fission of the target and recorded only fragments originating from nuclei that fission after recoiling out of the target. The inside surfaces of the chamber are covered with 99,999% pure Al-foils of 45 u thickness. Typically, the irradiation times varied between 16 and 24 hours per exposure. The proton flux was monitored with Al-foils.

After irradiation the mica detectors were annealed at 420°C for 6 hours to remove some of the very short tracks originating from spallation products$^{9)}$. The mica was etched in 48% HF at 25°C for 20 minutes. The detectors were scanned with an optical microscope at a magnification between 660 and 1250 fold. Only those tracks with projected track lengths greater than 4,5 u were recorded as fission fragments.

Scanning revealed tracks in both planes of the mica. Since one plane, here called the lower plane, was not exposed to fragments from isomeric fission, these tracks resulted from fission of heavy element impurities in the mica. Both planes were scanned and the number of tracks in the lower plane was substracted from those in the upper plane. Typically, 8 strips of mica, 0,5 mm wide and approximately 3 cm long were scanned. The strips were taken at 45° intervals.

The scanning results are summarized in Table 3. The fission fragment track density is given by the number of tracks divided by the area scanned in cm^2 and the numbers of protons in milli= coulomb. For the majority of the irradiations with UF_4 and ThF_4 targets, the track densities were somewhat higher than the average background (Al-blank); however, the results could not be reproduced consistently. This is due to the transfer of target material onto the Al-exit window 3 cm downstream from the target. The detectors register fragments from prompt fission of transferred material, as shown by the following experiments. The chamber covers from previous UF_4 and ThF_4 irradiations were transferred to clean chambers with an Al-foil in place of a target. The track density from the irradiations with used covers ("dirty blanks") was usually higher than for the normal blanks with a clean cover. Furthermore, the highest track density was obtained with an UF_4-target and a chamber cover from a previous UF_4-exposure (experiment Nr.6, Table 3).

Due to this transfer of target material, the closed chamber (Figure 4) was considered not suitable for this isomer search.

Therefore a second series of experiments was initiated. In order to prevent a constribution from prompt fission of transferred target material, irradiations were carried out in a vacuum chamber open at both ends. This geometry, shown in Figure 5, was located in the center of the 20 m long ISOLDE beam tube[8]). The target-detector geometry is the same as in Figure 4. Additionally, some experiments were carried out with a target-detector distance of 1 mm. There is no window to collect sputtered target material or isomers with half-lives longer than approximately 1 usec.

The results for irradiations using this geometry are given in Table 4. The background is one order of magnitude lower than that for the closed chamber. The track densities for the UF_4 , ThF_4 and Bi runs are not higher than the background within experimental uncertainties.

The result, that no isomers are seen in an "open chamber" (Fig.5) is in agreement with recent findings of Alexander[3]).

It is rather difficult to estimate an upper limit for the production cross section since there are many unknown factors such as: the velocity of the recoil nuclei in the target (i.e. the effective target thickness), the angular distribution of the recoil nuclei, and the detection efficiency. An estimate of the upper limit of σ was made by assuming that all fission events take place at a point source 2 cm from the center of the target and that the average recoil range in the forward direction is approximately 30 $\mu g/cm^2$. Given these assumptions, the upper limit for the production of spontaneously fissioning isomers from UF_4 , ThF_4 , and Bi is approximately $\sigma \leq 10^{-30}$ cm^2. In order to get a rough estimation of the half-lives that could be detected, it was assumed that the fissioning isomer moves with the average

Table 3: Experimental Results for Closed Chamber

Exp. No.	Target	Number of Protons x 10^{-16}	Number of Tracks [a]	Number of Fields [b]	$\frac{\text{Tracks}}{\text{mcoul-cm}^2}$
1	Al Blank	1.19	55 ± 10	378	30 ± 5
2	Al Blank	1.61	29 ± 7	324	14 ± 3
				Average	22 ± 6
3	UF_4(.500mg/cm^2)	2.01	44 ± 8	279	20 ± 4
4	Al Blank [c]	2.74	126 ± 13	110	104 ± 11
5	UF_4(.500mg/cm^2)	2.04	157 ± 13	57	336 ± 28
6	UF_4(.500mg/cm^2) [c]	.98	671 ± 26	209	821 ± 32
7	UF_4(.500mg/cm^2)	1.61	286 ± 17	477	93 ± 6
8	Al Blank [c]	1.56	78 ± 9	373	33 ± 4
9	ThF_4(.345mg/cm^2)	2.13	176 ± 14	162	127 ± 10
10	ThF_4(.345mg/cm^2)	2.32	586 ± 26	207	369 ± 16
11	ThF_4(.345mg/cm^2)	1.63	97 ± 11	445	33 ± 4
12	Al Blank [c]	1.35	297 ± 18	393	137 ± 8
13	Bi (.319mg/cm^2)	1.43	42 ± 8	215	34 ± 7

a) Difference between the number of tracks in upper and lower plane.
b) The area of a field is 0.25mm^2.
c) Using cover from the previous irradiation (dirty blank).

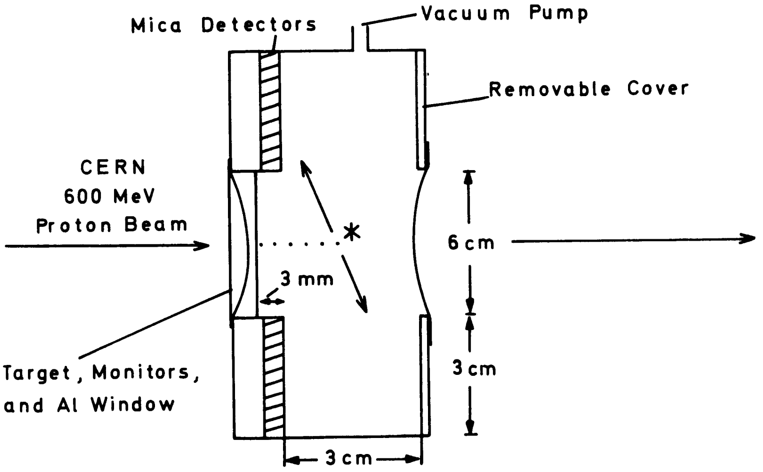


Fig. 4 Schematic drawing of closed chamber

* An isomer has left the target, decays in flight, and one fragment is registered in mica

Table 4: Experimental Results for Open Chamber

Target	Total Number of Protons x 10^{-16}	Number of Tracks [a]	Number of Fields [b]	$\frac{\text{Number of Tracks}}{\text{mcoul-cm}^2}$
Al Blank [c]	1.54	16 ± 10	812	3.2 ± 2.0
Al Blank [d]	2.49	7 ± 7	471	1.5 ± 1.5
UF_4(.451mg/cm^2) [c]	2.83	2 ± 4	822	0.3 ± 0.6
UF_4(.451mg/cm^2) [c]	2.16	4 ± 8	917	0.5 ± 1.0
UF_4(.451mg/cm^2) [d]	1.42	9 ± 7	473	3.3 ± 2.6
ThF_4(.626mg/cm^2) [c]	3.03	6 ± 19	921	0.5 ± 1.7
ThF_4(.626mg/cm^2) [d]	1.41	0 ± 7	409	0 ± 3.0
Bi (.700mg/cm^2) [c]	1.74	0 ± 5	765	0 ± 0.9
Bi (.700mg/cm^2) [d]	8.28	3 ± 5	414	0,2 $\pm$ 0.4

a) Difference between the number of tracks in "upper plane" and "lower plane" Due to the subtraction of the number of tracks in the lower plane from the tracks in the upper plane, the uncertainty is larger than the net effect.

b) The area of a field is 0.25 mm^2.

c) Distance from target to detector is 3 mm.

d) Distance from target to detector is 1 mm.

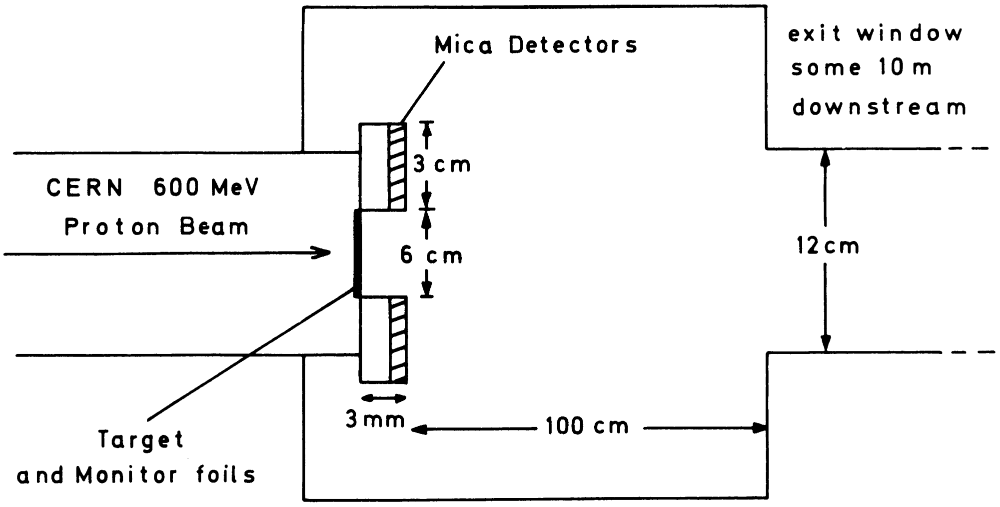


Fig. 5 Schematic drawing of open chamber

forward momentum imparted by 600 MeV protons on heavy target nuclei. Its velocity would be appr. 6 mm/10 nsec according to Porile[10]. This implies that half-lives in the order of 10 nsec would be detectable with this system, as quoted in Table 2 and Figure 1. It should be pointed out that these experiments do not rule out the possibility of the production of fission isomers with 600 MeV protons on U, Th, and Bi. If, for example, the isomers are produced by the interaction of protons on the nuclear surface, the momentum transfer in the forward direction may be so small that the short lived isomers would decay before recoiling the 1-3 mm necessary for detection.

IV. Acknowledgements.

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