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Discovery and cross-section measurement of 58 new fission products in projectile-fission of 750-A MeV ^{238}U .[★]

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The projectile fission of uranium at relativistic energy impinging on a Be target was investigated with the fragment separator, FRS, in order to produce and identify new isotopes and to measure their production yields. Fifty eight new fragments have been observed with a neutron excess of $(N-Z)/Z$ reaching 0.8 and cross sections as small as 0.3 nb.

In the field of nuclear structure, the exotic nuclei far from the line of β -stability attract a renewed interest. The neutron-rich side is particularly exciting since new phenomena have been predicted. Due to the weakening of the Coulomb field on the neutron-rich side, the neutron excess can be large and neutron matter at low density, as compared to neutron-stars, is expected to generate interesting nuclear structure effects; besides neutronic halos, the quenching of spherical shells is predicted by self-consistent mean field theories and even the double magicity of ^{78}Ni has been questioned [1]. From the astrophysical aspect, natural mass abundances related to the r-process allow tests of nuclear structure in the close vicinity of the path of this process. The shell quenching and neutron-skin effects are also called for in order to improve the agreement with sidereal mass abundances correlated with isotopes very far from stability [2].

[★] This work forms part of the PhD thesis of C. Engelmann

Progress in the understanding of nuclear deformations came mainly from the spectroscopy of fission products. It can be illustrated by the region of prolate deformation discovered in 1970 around the neutron-rich isotope of ^{98}Sr [3,4]. The region was explored by measuring β - γ decays after fragment separation; Thermal neutron induced fission revealed the accumulation of isomers at $N=59$ for elements in the vicinity of Zr [5] and allowed the detailed spectroscopy of a number of n-rich nuclei. More recently with the IGISOL technique, the transitional region of $^{108-114}\text{Ru}$ and $^{106-108}\text{Mo}$ was investigated with p-induced symmetric fission of ^{238}U [6].

A qualitative step has been accomplished in the meanwhile with the use of big γ detector arrays observing spontaneous fission of ^{252}Cf [7] and ^{248}Cm [8]. With ^{248}Cm new neutron-rich fragments between Sr and Mo were discovered and studied. However, β - γ spectroscopy remains complementary to γ - γ studies and it is an experimental challenge to find new efficient ways to get access to very neutron-rich nuclei.

Our first experiment with fission of relativistic Uranium-ions on a lead target demonstrated that fission in inverse kinematics is powerful to produce and observe neutron-rich species [9]. Fifty-three new fission fragments were identified in this experiment. The fission process, observed over a large range of excitation energies, was investigated in parallel. The low excitation energy regime is dominated by the Coulomb fission of the uranium projectile [10,11], whereas fission of highly excited uranium residues is induced by more violent collision with the target nucleus [12]. In this letter we report on the 58 new neutron-rich isotopes discovered in the region of elements between Ca and In and on the first determination of their production yields.

The experimental method, described in previous papers [9,10] is briefly summarized here. Uranium ions extracted from a Penning source were preaccelerated in the UNILAC. They were injected into the heavy-ion synchrotron, SIS, and extracted at an energy of 750 A MeV. Fission fragments were separated using the fragment separator FRS [13] tuned in an achromatic mode and identified by event-by-event measurements of ΔE - $B\rho$ -ToF and trajectory. Three to five isotopes of twenty elements were transmitted simultaneously. The 1% accuracy needed on the energy-loss measurement ΔE for element identification was achieved by using the ionization chamber MUSIC [14] with a continuous control of the gas pressure. With the nuclear charge Z determined that way and by knowing that $q=Z$, the mass identification was carried out by measuring the time of flight (ToF) and the magnetic rigidity $B\rho$ with an accuracy of 10^{-4} . The length of the trajectories were corrected for their angle, measured at the exit of the FRS. The calibration in mass A and nuclear charge Z was derived from the uranium primary beam and confirmed by the enhanced yield of ^{134}Te in low-energy fission. Each FRS-setting provided the count-rate of identified isotopes at a defined value of $\beta\gamma$, the momentum/nucleon.

Fission fragments emitted in forward direction were selected by setting the FRS at a magnetic rigidity larger than the projectile rigidity. The high velocity comes from the relativistic addition of projectile and center of mass fission velocities in forward direction. The large A/Z ratio imply a very peripheral reaction process, in which no or only a few neutrons are evaporated; therefore the selection of large rigidity fragments acts as a trigger on fission at low excitation energies [10,11]. In the U-frame fission velocities, β_f , are determined by Coulomb repulsion at scission. For each Z_1 - Z_2 partition they are given by $\beta_f=f(Z_1)$, decreasing from 0.055 for Ni to 0.035 for Ce. This velocity dependence on Z_1 is assumed to be the same as in fission induced by thermal neutrons[15]. The validity of this assumption was demonstrated by deriving the fission velocity from the measured distribution of laboratory velocities for fission fragments emitted forward or backward [11,12]. In the laboratory, the $\beta\gamma$ distributions are -to first order- the same for all isotopes of a given element. Thus for fission fragments, the increase of $B\rho$ scales the increase of A/Z .

After the first experiment with the uranium-beam in 1992, the intensity was increased by two orders of magnitude up to $2 \cdot 10^7$ ion/s. Therefore a region of new isotopes with larger neutron excess became accessible. Beryllium and lead targets were compared for this purpose. For similar thicknesses of 1. and 1.2 g/cm² respectively (i.e. similar energy losses), the beryllium target includes twenty times more atoms/cm² than the lead target. They both lead to equivalent target location straggling effects and thus to the same momentum widening. Since the cross section for low energy fission in Be was measured to be 120 mb, i.e. 18 times less than in Pb [10], the fission rates are about the same for both targets. However, yields of both symmetric fission with $Z_1 \approx Z_2$ and very asymmetric fission with Z_1 down to 24 were found to be larger on Be than on Pb; the Ni yield for example was measured to be ten times larger on Be than on Pb. Since the width of the isotopic yield distributions are rather similar for nuclear as for Coulomb fission [11], the Be target was chosen to identify new light fission fragments.

Starting from a rigidity $B\rho = 1.05 \cdot B\rho_0$, where the maximum of the fission yield has been found, the rigidity was increased step by step up to $B\rho = 1.16 \cdot B\rho_0$ providing a scan of production yields for increasing masses. Although the yields are dropping by roughly a factor of eight per extra-neutron, the count-rates were found still large enough for isotope identification. The frontier depends mainly on the beam-intensity since the background rate becomes extremely low at large magnetic rigidities. An effective luminosity of $2 \cdot 10^{28}$ cm² s⁻¹ was reached at the FRS for this experiment. This is approximately two orders of magnitude more than in the investigation of Ni at Lohengrin using thermal neutron induced fission [16]. The simultaneous observation of twenty elements in the same range of A/Z renders the measurement very efficient, in particular for determining relative yields.

At very low counting rates and large rigidities, background from atomic charge exchange processes in the intermediate scintillator, in the windows of the FRS vacuum or of the MUSIC detector cannot be totally ruled out. A second measurement of the energy-loss was made in the intermediate focal plane via a thin mini-ionization chamber. This double check on Z eliminates any contamination from atomic processes. We could indeed verify that those processes were negligible for the light fission products.

At a rigidity of $B\rho = 1.16 \cdot B\rho_0$, a three day accumulation was performed during this second experiment with a total dose of 10^{13} U-ions on the Be target. Fission products with A/Z as large as 2.78 were selected, the second half of the FRS being tuned on Ni. In Fig. 1 are shown mass distributions of all elements from Ti to Nb, obtained in this measurement. They are not corrected for the variations of transmission through the FRS. In each spectrum the transmission decreases for $A/Z < 2.7$, since the ions start to run apart from the optical axis. Comparing elements, the transmission is reduced for the large atomic numbers by the Z -dependent energy losses in the start-scintillation detector located in the dispersive focal plane. This reduction is clearly seen on Fig.1 for Nb at the end of this Ti to Nb series.

Out of this measurement, the observation of six new nuclei, ^{80}Cu , ^{77}Ni , ^{78}Ni , ^{73}Co , ^{74}Co and ^{75}Co was already reported [17] and preliminary data on other isotopes were shown at conferences on exotic nuclei [18] and on radioactive nuclear beams [19]. Because of electronic thresholds, the spectra of the elements Ca and Sc, plotted in the bottom-left of Fig.1, were obtained with a lower efficiency than for the other elements. On top of the figure the Mo to In spectra were measured with a different FRS setting, $B\rho = 1.12 \cdot B\rho_0$, and a U-dose of 10^{12} U-ions. The discovered isotopes are marked on the figure by their mass numbers. In parenthesis are indicated isotopes found in other experiments between the first (1992) and the present measurement. From the systematics of the data, taken at different magnetic rigidities [20], the cross sections were calculated. In Table 1, the 64 new isotopes, including Cu, Ni and Co are listed with their production cross sections. Values as small as 0.2 nb were measured for the most neutron rich isotopes with $A/Z = 2.78$. Statistical errors are the main source of uncertainties for fragments produced with low rates. Systematic errors, due to the evaluation of the FRS-transmission and to the determination of the beam intensity, limit the accuracy on the whole set of numbers to 30%.

In the course of our two experiments at the FRS, in 1992 on Pb [9] and now on Be, altogether 117 new isotopes were identified. They are indicated by points on the chart of nuclei (see Fig. 2). Five to six new isotopes are found for each element in the fission valley. It is explained by the filling of the valley, already observed in our study of U+Pb collisions. Furthermore, in the valley the rates per element were found to be three times larger on the Be-target than

on the Pb-target [10]. From Ni down to Ca three new isotopes per element are identified. Starting from Zr, the elemental yield decreases with Z. it is minimum for Ca and increases again rapidly for light elements in agreement with many previous results of p+U studies [21].

Even though all neutron-rich nuclei discovered here remain quite far from the neutron drip-line, they approach the r-process path. The physical conditions of the r-process were recently revisited [22]; instead of starting on stable Fe isotopes, residues of fusion, the neutron capture would start on much lighter nuclei after the destruction of Fe by a neutrino wind which accompanies the Super-Novae. The new site proposed does not rule out the part played by the neutron shell closures at $N=50$ and 82 . On Fig.2 is shown the r-process path recently calculated by P. Möller and K.-L. Kratz [23]. The new isotopes are reaching the path between Ni and Br and between Cd and Te. The distribution of mass-abundances calculated by these authors agree with the observations except for two regions. In the most troublesome, between $A=110$ to 125 , the predicted abundances are one order of magnitude below those observed. In this region nuclear constants can now be measured. It was already demonstrated at the FRS that for very n-rich fragments, in spite of low counting rates, halfives can be obtained [24] [25]. Binding energies become accessible by mass measurements at the ESR [26].

The present findings open a way to future investigations of nuclear properties of importance for the r-process in nucleosynthesis. Besides, less neutron rich isotopes are produced with cross sections or rates which may be sufficient for β -n or even β - γ spectroscopy. Such studies would carry on the investigations on shell closures at $N=28$ and 50 and on deformation around ^{100}Sr as already quoted. With projectile fission of U-ions a new region of more than a hundred isotopes between the doubly magic nuclei ^{132}Sn and ^{78}Ni is now accessible for spectroscopic investigations.

Figure Caption

Fig. 1: Identification spectra of the new isotopes after separation with the FRS. From Ti up to Nb data were collected in a single long measurement at $B\rho = 1.16 (B\rho)_0$. The detection efficiency of Ca and Sc was lower, as can be seen in the spectra in the bottom left corner. Elements beyond Nb were measured with a lower rigidity for a shorter time. New isotopes are indicated here by their mass numbers.

Fig. 2: Location of the new isotopes on the nuclear chart. The r-process path [23] is shown by the thick line and the limit of nuclear binding is shown by the dashed line.

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Isotope	Counts	σ (nb)	Isotope	Counts	σ (nb)	Isotope	Counts	σ (nb)	Isotope	Counts	σ (nb)
⁵⁴ Ca	11	5	⁶⁸ Mn	43	5.0	⁸⁸ Ge	67	6.7	¹⁰⁴ Sr	72	6.6
⁵⁵ Ca	6	2	⁶⁹ Mn	5	0.4	⁸⁹ Ge	11	0.6	¹⁰⁵ Sr	8	0.8
⁵⁶ Ca	3	1	⁷⁰ Fe	200	28	⁹⁰ As	228	21	¹⁰⁶ Y	112	16
⁵⁶ Sc	68	50	⁷¹ Fe	39	4.7	⁹¹ As	37	3.2	¹⁰⁷ Y	21	2.8
⁵⁷ Sc	30	10	⁷² Fe	2	0.3	⁹² As	4	0.6	¹⁰⁸ Zr	336	100
⁵⁸ Sc	11	3	⁷³ Co	154	20	⁹² Se	1061	120	¹⁰⁹ Zr	47	5.4
⁵⁹ Ti	115	50	⁷⁴ Co	18	2.0	⁹³ Se	117	8.2	¹¹⁰ Zr	5	0.4
⁶⁰ Ti	40	10	⁷⁵ Co	2	0.3	⁹⁴ Se	31	2.0	¹¹¹ Nb	150	50
⁶¹ Ti	9	2.5	⁷⁷ Ni	12	1.4	⁹⁵ Br	781	75	¹¹² Nb	22	2.5
⁶² V	104	21	⁷⁸ Ni	3	0.2	⁹⁶ Br	69	5.9	¹¹³ Nb	8	0.7
⁶³ V	28	6.4	⁸⁰ Cu	6	1.0	⁹⁷ Br	8	0.7	¹¹⁴ Mo	8	24
⁶⁴ V	2	0.3	⁸² Zn	125	13	⁹⁷ Kr	2110	500	¹¹⁶ Tc	29	81
⁶⁵ Cr	82	7.8	⁸³ Zn	10	1.0	⁹⁸ Kr	525	54	¹¹⁷ Tc	3	8.4
⁶⁶ Cr	19	1.5	⁸⁵ Ga	61	6.5	⁹⁹ Kr	32	2.6	¹¹⁹ Ru	5	14
⁶⁷ Cr	4	0.5	⁸⁶ Ga	4	0.6	¹⁰⁰ Kr	3	0.5	¹²² Rh	5	13
⁶⁷ Mn	245	40	⁸⁷ Ge	583	67	¹⁰³ Sr	409	50	¹²⁴ Pd	19	32

Table 1: Production cross section of neutron-rich fission fragments in the reaction 750-A MeV ²³⁸U on Be. The number of counts refer to the sum over all different FRS-settings. Light elements were observed at $B\rho / B\rho_0 = 1.16$ whereas the heavier elements were seen at 1.14 and at 1.12.