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REACTION AND FISSION CROSS-SECTIONS OF 750 A·MEV ^{238}U IONS ON Pb, Cu AND Al-TARGETS

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Abstract; Charge-loss and fission cross-sections of ^{238}U at 750 A·MeV were measured on Al, Cu and Pb targets. The charge-loss rate was obtained by the attenuation method. Fission was selected by detecting the pair of highly ionizing fragments. Since the neutron-loss cross sections were measured in a parallel experiment for the same projectiles, all cross sections contributing to ^{238}U collisions on nuclei are available now as function of the target mass number and can be compared with current models.

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Introduction

The acceleration of Uranium ions to relativistic energies by the SIS at GSI-Darmstadt opens a new area for fragmentation and fission studies. Using inverse kinematics, the efficiency for detection of the fragments is multiplied by several orders of magnitude compared to target fission or spallation studies since reaction products are forward focussed in a narrow cone with large laboratory velocities. With the fragment separator FRS, Uranium projectile fragments [1] [2] and fission fragments [3] were separated and isotopically identified in flight by the $B\rho$ - ΔE -ToF method. The elemental yields deduced for fission are reported in a following publication in this journal.

Fission observed in collisions of Uranium projectiles with target nuclei is due both to electromagnetic and to nuclear processes. At large impact parameters, the long-range Coulomb force prevails. The projectile is excited mostly to the giant resonance region by an exchange of virtual photons; it then decays by neutron emission or by fission. For smaller impact parameters, peripheral nuclear collisions take place and the fissile projectile is left abraded and excited. After de-excitation by nucleon emission, it can undergo fission with a finite probability. In an inclusive fission measurement, as the one we report here, the cross section results from the addition of multiple chance fission processes.

During the first Uranium beam experiment at FRS in November 92, inclusive measurements of fission and charge exchange reactions were performed for 750 A·MeV ^{238}U projectiles

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on Pb, Cu and Al targets located behind the FRS. After a selection of a U^{92+} beam by the FRS, two ionization chambers located in front and after the target were used to identify Uranium ions and fission products. Our experimental results are compared with data from a previous measurement, performed with the 1 A·GeV ^{238}U beam delivered by the BEVALAC [4]. More recently, the electromagnetic dissociation of ^{238}U was investigated also at the BEVALAC at 120 A·MeV incident energy [5].

Eventhough neutron abrasion or evaporation of U do contribute to the reaction our measurement is insensitive to neutron-losses. The few-neutron removal cross-sections (^{238}U , $^{238-xn}\text{U}$) were measured in our energy domain [6] during the same beam time. Total reaction cross sections could be obtained by adding charge-loss to neutron-loss cross sections. They are compared here with the predictions of strong interaction processes which are either mere parametrizations of total nuclear reaction cross-sections [7], [8] or Glauber-type calculations [9].

Results are relevant to accelerator boosted reactor technology in order to anticipate the isotopic composition of the nuclear residues created in the primary process of the collision of 1 GeV proton beam with fissile target nuclei.

Experimental set-up

The ^{238}U beam of 2×10^5 ions was delivered by the SIS in the ionic charge state 73^+ . The ions were partially stripped by a thin copper foil (205 mg/cm²) placed at the target position of the fragment separator FRS [10]. Ionic charge states were separated with the FRS and U^{92+} ions were focussed on the target at the final image plane of the separator. At the exit of the FRS, two four stage Multiple Sampling Ionization Chambers [11], located before and after the target (MUSIC1 and MUSIC2) were used to identify the nuclear charge. With MUSIC1 the primary U ions were counted. The MUSIC2 was used either in a standard mode in order to measure the attenuation of the primary beam or as a Time Projection Chamber (TPC) to enumerate fission events from the detection of a pair of ionizing fragments [12](see Fig. 1).

Fission fragments are emitted back-to-back isotropically in the projectile frame. The transfer of momentum between the target nucleus and the U projectile can be neglected [13] and the fission kinematics is assumed to be the same as for an in-flight decay taking place in the target. The phase space of momentum vectors, spherical and homogeneous in the center of mass system of ^{238}U converts in the laboratory into a forward cone of approximately 70 mr angular aperture, its rotational axis being the projectile direction. The end points of the momentum vectors lie on an ellipsoidal shell. The momentum of each fragment of a pair remains correlated to its partner. The ellipsoidal shell has a non-zero thickness due to different mass partitions and to the distribution in total kinetic energy given by fission.

In the previous experimental work of Greiner [4], the identification of fission events was based on the measurement of the sum of the fragment energy-losses in Si detectors. The velocities of fission fragments remain close to velocity of the U-projectiles. Therefore their energy deposit is only a function of the square of their atomic number Z and the spectrum of the sum of the energy losses for a pair of fragments shows a single peak close to half of the energy loss of U-projectiles.

In our work, fission events were identified from the signals of each of the two fragments detected in the MUSIC2 used as a TPC. The electrons created along the fragment trajectories, were accelerated by an horizontal electric field perpendicular to the beam and collected on four vertical anodes shielded by a Frisch grid (see fig. 1). Both signals were processed

independently. Fission fragments could be sorted only when the time difference between the electron drift times for the two fragments of a pair was measurable, i.e. larger than 3 μ s. When a pair of fragments was emitted close to the plane perpendicular to the electric field containing the beam direction, the difference between the electron drift times of the two fragments was too small and only one signal was processed instead of two. A dead region of space results from this limitation which introduces an inefficiency in the detection of approximately 20%. It was evaluated by using two large plastic scintillation detectors located behind the chamber and divided by a narrow horizontal slit in order to let the beam through [12].

The distance between MUSIC2 detector and target was optimized for fission measurements; it was small enough to detect all fission fragments emitted within a forward cone of 70 mr angle and large enough to minimize the dead angle for which fission products cannot be separated. The contribution of residual matter between the detectors (detector windows, a multi-wire chamber and 1.2 meter of air at atmospheric pressure) was evaluated by a target-out measurement. It corresponds to almost 50% of the target.

Results

a) Charge loss cross sections

The ratio of the U ions measured before and after the target provides the charge-changing cross section (see Fig. 2). There is no significant loss of U ions by elastic or inelastic scattering since both processes are negligible for angles larger than 70 mr. The capture of nuclear charges has a small probability compared to charge-loss ($\sim 10^{-4}$) (see Fig. 2b) and can be neglected. The charge changing process defined here includes fission, fragmentation and central collisions. It stands for the total reaction minus all non-charge changing reactions.

The structure in the energy-loss peak of U ions detected by MUSIC1 (Fig. 2a) clearly indicates the contribution from the three atomic charge states, 92^+ , 91^+ and 90^+ . Bare U ions, rigidity selected in the FRS, capture electrons when they cross thin foils of matter. Atomic charge equilibrium is established when the beam passes through the exit window of the FRS (218 mg/cm² Al equivalent) before the entrance into the MUSIC1. The probability for further ionic charge exchange to take place in the MUSIC2 detector is small, given the small thickness of the filling gas ($t = 90$ mg/cm² of Ar+CH₄) and therefore ionic charge states remain mostly unchanged in the MUSIC detectors.

The energy loss spectrum of U ions (Fig. 2b) measured in MUSIC2 shows a single wide peak for U, since the ΔE resolution is not as good when the chamber is used in a TP-mode. Even if the energy resolution were perfect, nuclear charge-loss by one or two units could not be disentangled from atomic charge exchange by measuring only ΔE . Therefore we can only evaluate cross sections for the loss of more than two nuclear charge-units, i.e. $\sigma_{\Delta Z > 2}$.

At smaller energy losses, signals from less ionizing fragments are seen on the left of the U peak in Fig. 2b. They arise from projectile fragments with lower nuclear charges and almost unchanged velocities.

The charge-loss cross section in the target, $\sigma_{\Delta Z > 2}$ (or σ_t) was calculated as follows ;

$$n_2/n_1 = \exp[-(\sigma_t x_t + \sigma_b x_b)]$$

where n_2/n_1 is the ratio of the number of ions with q (or Z) = 92, 91 or 90 detected in

MUSIC2 versus the number of U ions counted in MUSIC1 in any of the three atomic states. x_t is the target thickness in $\text{atoms} \times \text{cm}^{-2}$. The index b refers to the same quantities for target-out counting.

Since the targets are thin, the ratios n_2/n_1 are close to unity, and the “thin target” approximation is valid ; therefore $n_2 = n_1 (1 - \varepsilon)$ with $\varepsilon = \sigma_t x_t + \sigma_b x_b$

Data from target-out measurements yield $(n_2/n_1)_b = 1 - \sigma_b x_b$

The values of σ_t are listed in Table 1. In spite of good statistics the uncertainty reaches 10 % due to a large target-out contribution. A good agreement is obtained with charge-loss cross sections previously measured by Greiner at 1 A.GeV [4], see Table 1. Values for Pb, Cu and Al are extrapolated down to Be, assuming that charge loss is mostly due to nuclear interaction. Then the cross sections are proportional to the square of the strong absorption radius i.e. $(A_p^{1/3} + A_t^{1/3})^2$ [14].

b) Fission cross-sections

Fission events were sorted by different methods.

1) Counting the events for which two fragments of intermediate Z were detected in MUSIC2.

2) Measuring the energy loss of the two fragments. The time differences between the signals of both fission-partners were measured in the forth section of the MUSIC2 detector (Fig. 3). They provide the distance between the trajectories in the last stage of the detector with an optimum accuracy. Due to equality (in the center of mass) and to conservation of perpendicular momenta of the two fragments from center of mass to laboratory system, this distance is proportional to the perpendicular component $p_\perp$ of the fission fragments momenta projected along the electric field. For a unique type of partition (Z_1, Z_2) occurring at a fixed kinetic energy, a rectangular shape should be observed. However, the small time differences are suppressed because of the “dead-region”. At the largest time gap a Gaussian fall off is observed since there is a broad distribution in masses and nuclear charges as well as in total kinetic energy in fission.

The time spectrum (Fig. 3) was linearly extrapolated towards very small time differences and integrated to obtain the number of fission events. The proportion of undetected pairs of fragments was found to be 20 %, consistently with the evaluation from the scintillator detectors.

3) The spectrum of the energy loss for the first fragment of a pair was analyzed, as shown on Fig. 4b. Three wide peaks are seen: Low ΔE signals are due to light fission fragments and intermediate ΔE to heavy fragments. The third peak at larger ΔE comes from pairs of fragments for which the two unseparated pulses are added. Each fission leading to one, and only one count, fission events can be enumerated. The scatter-plot of the energy losses of correlated fragments (Fig. 4a) confirms the attribution of the first two peaks. It shows also the asymmetry of the fission fragment elemental distribution. Note that “hot” fission events can be seen in the valley. For a given ΔE (or Z), the width of the Z range of the partner is larger in the valley than for the asymmetric break-up. Indeed while the Z sum is well defined to 92 in the asymmetric region, fission of U fragments (with $Z < 92$) can be observed in the valley.

Target-out contributions were subtracted with the three methods. It reduces the accuracy of the measurement. Together with the uncertainty due to the estimation of unresolved pairs of fragments, the precision ranges from 7 % to 10 % according to the target.

The three methods lead to mutually compatible fission cross sections. The weighted mean

values are given in Table 2. They are found to be larger by 30 % to 50 % than in the work of Greiner et al. [4] performed at a slightly higher energy (1 A.GeV). As mentioned already these authors used only the energy loss spectra in which they could not discriminate events with two fragments (fission) from those with one highly ionizing fragment. The background continuum due to fragmentation-type events might have been overestimated. In the present work, we further required that a pair of fragments must be found, each with an appropriate energy-loss.

Another recent measurement of Pb+U target-fission cross sections [13] energy interpolated to 750 A.MeV yields a value of (3.2 ± 0.4) b, consistent with the present result of (3.54 ± 0.21) b.

Discussion

At relativistic energies, U projectiles interact with target nuclei by Coulomb forces and, for impact parameters b smaller than b_{min} , by nuclear force. The total cross section can be decomposed into a sum of nuclear and electromagnetic (EM) amplitudes:

$$\sigma_{tot} = k[A_{nucl} + A_{em}]^2$$

where k is a constant. An interference term between the nuclear (A_{nucl}) and the Coulomb (A_{em}) transition amplitudes may occur which is not necessary negligible in case of highly charged nuclei. The GDR, for example, may be excited by either the nuclear or the Coulomb field and decay via fission or xn emission. In the case of collisions of U on Au at 960 A.MeV, Benesch et al. [8] have calculated that 13 to 25 mb would correspond to this interference term while they obtained 4.1 b for the EM-cross section. For lighter systems this contribution is even smaller. Therefore given the present accuracies in measurement, this interference term can be neglected. and in the following we use

$$\sigma_{tot} = \sigma_{nucl} + \sigma_{em}$$

The values of b_{min} are parametrized as a function of the radii of the colliding nuclei by Kox [7] or by Benesh [8]. In this second model -BCV- the limiting impact parameter is written as,

$$b_{min} = 1.34 \cdot [A_p^{1/3} + A_t^{1/3} - 0.75(A_p^{-1/3} + A_t^{-1/3})]$$

The assumption of two additive contributions is extended to fission for which

$$\sigma_f = \sigma_{nucl,f} + \sigma_{em,f}$$

The EM-fission, function of kinetic energy and of atomic number of the target, becomes negligible in case of a Be target. This is confirmed by the recent work of Rubehn [15] where the EM-fission has been measured at 600 and 950 A.MeV using two selection criteria:

- 1) by requiring that the sum of the fragment atomic numbers be 92 and
- 2) by excluding events for which a charged particle emitted from the interaction zone would be detected. The results interpolated in energy to 750 A.MeV are shown on Fig. 5. The value obtained for the "low-energy" fission component separated at the FRS on the Pb target by triggering on fragments with momenta larger than projectile momentum [16] is indicated for comparison.

The experimental results for σ_f , $\sigma_{nucl,f}$ and $\sigma_{em,f}$ are plotted in Fig. 5. They show to what extent nuclear interaction is responsible for fission. Indeed, in our experiment most of the so-called fission events originate from fission of projectile-fragments of U, produced by neutron-evaporation or proton-abrasion.

The total *charge-loss* cross sections, $\sigma_{\Delta Z}$, are obtained by adding the calculated cross sections for the loss of one and two nuclear charges, $\sigma_{\Delta Z=1,2}$, to our data for $\sigma_{\Delta Z>2}$. The former are calculated using the up-dated version of the statistical abrasion code of Gaimard and Schmidt [17] (see Table 2 second row) which was proven able to reproduce few-nucleon removal cross sections with a good accuracy [1]. Fragmentation is calculated by subtracting the values of fission from the total charge-loss results (see Table 2, row 5). The central collisions, analyzed in the work of Greiner [4], occur with a small probability. They are presently included in the fragmentation channel.

In order to determine the electromagnetic cross sections, multi-neutron removal and the electromagnetic fission contributions are added.

$$\sigma_{em} = \sigma_{em,f} + \sigma_{xn}$$

The xn removal from ^{238}U was measured recently at energies of 600 and 950 A·MeV for a series of target nuclei [6]. Values interpolated to the energy of 750 A·MeV are reported in row 6 of Table 2. Removal of more than 4 neutrons can be neglected. Results obtained by the sum above compare well with the values calculated by Aumann [6] on the basis of the formulation of Bertulani [18].

The *total reaction* cross sections are given by the sum of charge-loss and neutron-loss cross sections, $\sigma_{tot} = \sigma_{\Delta Z} + \sigma_{xn}$.

Experimental results are reported in the last line of Table 2. All values entering in the sum are taken from experiments except for the 1p and 2p-removal contribution which represents 3 % (or less) of the process. Fission, fragmentation, xn partial cross sections and their sum, the total reaction cross sections, are plotted in Fig. 6 as a function of the target mass number.

It is worth noting that processes ending in fission correspond to less than a third of the total cross-section. The xn contribution grows rapidly with the target atomic number since it originates mainly from electromagnetic interaction (see Fig. 5). On the other hand, total fission and fragmentation cross sections increase more slowly with the target mass (see Fig. 6).

The total cross sections are compared with model calculations. They are given by the sum of an electromagnetic contribution calculated as described above and the nuclear contribution evaluated by the interaction of strongly absorbing spheres [7] expressed also as a function of b_{min} . Values of b_{min} from Kox and from BCV were used for this calculation.

A third evaluation of the nuclear contribution was obtained by using the model of T. Brohm [9], who improved the statistical abrasion model [19], introducing soft limits of the nuclear spheres and transparency due to Pauli principle.

On Table 3 the values of σ_{nucl} , calculated by subtracting σ_{em} from σ_{tot} are reported. They are found in good agreement with the nuclear cross sections calculated according to the three methods mentioned above (See also Table 3). With the BCV parametrization of b_{min} the differences with experimental values are only 1.3 % for Pb, 5 % for Cu and 7 % for Be, and the results given by the model of Brohm are even closer (2 % for Be to 3 % for Pb). As in the case of EM-fission [15], the Kox parametrization does not fit so closely the results for heavy targets. A surprising result from this study is that reactions induced with relativistic fissile ions lead to twice more proton-rich than neutron-rich nuclei.

This might be of relevance to hybrid fission-spallation reactor systems in case the synergetic accelerator-reactor systems as proposed first by G. Bartolomew in 1966 [20] and promoted for energy production and waste transmutation [21], [22], would be developed. The reaction of relativistic protons with actinide nuclei produces about 1500 different isotopes by fission and spallation. The deexcitation of these isotopes governs the neutron

production. Their yields and halfives determine the balance of incinerated and newly produced radioactivity in case of nuclear incineration. The relative cross sections of fission and fragmentation as measured here are decisive to optimize the hybrid fission-spallation reactor systems. In thin target experiments all yields for fission and spallation products could be investigated in inverse kinematics using ^{238}U , ^{232}Th or ^{208}Pb beams bombarding H targets. Production cross sections and β^- halfives of any reaction products could be investigated using the existing FRS-facility.

Conclusion

Reaction cross sections for collisions of relativistic ^{238}U projectiles on different targets were revisited with the beam delivered by the SIS at GSI. Charge loss results are in good agreement with previous results. Discrepancies are found in case of fission but the discrimination of fission events has been presently improved compared to the previous experiment. Using results of other parallel experiments performed at GSI, the share of EM and nuclear components could be evaluated for fission and for total reaction cross sections. The former is calculated using the parameters of giant resonances in U and the second is calculated using up-dated nuclear absorption models. The calculated values compare well with our data. However, further experimental work would be required to discriminate between the models. Important at this stage is the fact that neutron-deficient products are twice more abundant than neutron-rich in collisions of light charged particles on fissile target nuclei at relativistic energy. Moreover, our results may become relevant for accelerator driven power reactor technology.

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FIGURE CAPTION

Fig. 1 Schematic view of the experimental set-up

Fig. 2 Energy-loss spectra of U measured in the MUSIC chambers :

- a) before the target,
- b) after the target

Fig. 3 Spectrum of time differences between the two fission fragments measured in the last section of the MUSIC2 detector. The spectrum can be extrapolated to $\Delta t=0$ since a rectangular shape is expected (see text).

Fig. 4 a) Correlations of energy losses between the partners of a fragment pair. The asymmetry in charge distribution is clearly observed.

b) Energy loss spectrum of the first detected fragment from a pair. The first two maxima are caused respectively by the light and the heavy partners. The last peak is due to unseparated pairs of fragments.

Fig. 5 Total fission cross sections -full line- and EM fission cross sections -dashed line- of 750 A·MeV U projectiles on different targets. The result obtained from FRS data analysis is shown with a full square.

Fig. 6 The total reaction cross sections ($\blacktriangle$), can be subdivided into a) fragmentation channel ($\blacksquare$), b) fission channel ($\bullet$) and c) multi-neutron emission channel ($\blacklozenge$).

TABLE CAPTION

Table 1 Target thicknesses and results for charge loss measurements.

Table 2 Fission and fragmentation cross sections. The charge loss cross sections, including fission, added to multi-neutron emission cross sections gives the total reaction cross sections.

Table 3 The values from em dissociation [6] are subtracted from the total reaction cross-sections. The nuclear contributions resulting are reported with the values obtained from the quoted models.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

Fig. 5

Fig. 6

Table 1

Target	Al	Cu	Pb	foils + air: Al eq.
Thickness mg/cm ²	1119	780	709	477
ε	$129 \pm 5 \cdot 10^{-3}$	$94 \pm 2 \cdot 10^{-3}$	$86 \pm 2 \cdot 10^{-3}$	$55 \pm 2 \cdot 10^{-3}$
$\sigma_{\Delta Z > 2}$ in barn Greiner	4.20 ± 0.43	5.45 ± 0.46	9.69 ± 1	
$\sigma_{\Delta Z > 2}$ in barn	3.93 ± 0.34	5.36 ± 0.42	9.15 ± 1.0	

Table 2

Cross Sections in barn	U + Be (extrapolated)	U + Al	U + Cu	U + Pb
$\sigma_{\Delta Z > 2}$ exp.	2.90 ± 0.3	3.93 ± 0.34	5.36 ± 0.42	9.15 ± 1.0
$\sigma_{\Delta Z = 1,2}$ (Junghans)	0.14	0.16	0.16	0.20
$\sigma_{fission}$ exp.	1.03 ± 0.1	1.34 ± 0.09	1.95 ± 0.13	3.54 ± 0.21
$\sigma_{fission}$ Greiner		1.10 ± 0.13	1.27 ± 0.14	2.73 ± 0.20
$\sigma_{fragmentation}$	2.00 ± 0.40	2.75 ± 0.35	3.57 ± 0.44	5.81 ± 1.02
σ_{zn} [Aumann]	0.20 ± 0.03	0.40 ± 0.05	1.01 ± 0.15	4.06 ± 0.40
σ_{tot} exp.	3.24 ± 0.32	4.49 ± 0.35	6.53 ± 0.44	13.41 ± 1.0

Table 3

Cross sections in barn	U + Be	U + Al	U + Cu	U + Pb
σ_{tot} experiment	3.24 ± 0.32	4.49 ± 0.35	6.53 ± 0.44	13.41 ± 1.00
σ_{EMD} [Aumann]	0.03	0.24	1.00	5.62
σ_{nuc} experiment	3.21 ± 0.32	4.25 ± 0.35	5.53 ± 0.44	7.80 ± 1.00
σ_{nuc} [Kox]	3.26	4.63	6.22	9.52
σ_{nuc} [Brohm]	3.32			7.54
σ_{nuc} [Benesh]	3.43	4.42	5.51	7.95