# Peculiarities in fragment mass distribution in the $^{238}U + {}^{40}Ar$ (243 MeV) reaction

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**Abstract.** A pronounced fine structure (FS) in the form of distinct peaks was observed in neutron gated mass spectra from the decay of the <sup>278</sup>110 composite system produced in the reaction <sup>238</sup>U + <sup>40</sup>Ar (243 MeV) at an initial excitation energy  $E^* > 70$  MeV. The FS peaks are located in the vicinity of mass numbers 70–80, 100, and 130, which correspond to those of magic nuclei (clusters). In the data there is also evidence for a new type of decay — collinear cluster tripartition of an excited nucleus.

**PACS.** 25.70.Lm Strongly damped collisions – 25.70.Pq Multifragment emission and correlations – 25.85.Ge Charged-particle–induced fission

## 1 Introduction

Recent experiments on the synthesis of superheavy elements [1-3] gave rise to a new wave of interest in shell effects [4]. In this field extraction of the production crosssection of compound nuclei at excitation energies of 15–30 MeV is very important but the necessary measurements are extremely time-consuming. However, it should be possible to study the essential features of the reactions leading to superheavy elements also at higher excitations when the fusion-fission cross-sections increases considerably. One of the goals of our experiment was to search for the manifestation of nuclear shells in the mass spectrum of the fragments from the reaction  ${}^{238}U + {}^{40}Ar$  at the bombarding energy  $E_{\rm Ar} = 243$  MeV. It was shown in earlier experiments [5–8] that in the interaction of heavy nuclei with heavy beams  $(A \simeq 40)$  at intermediate energies the mass spectra exhibit peaks suggesting a preference for asymmetric fragmentation ( $A_1 \simeq 90, A_2 \simeq 200$ ). Later experimental results [9] confirmed the prediction given in [10] that shell effects are noticeable in Z = 108 systems up to 50 MeV excitation energies of the compound nuclei. Apart from the problem of the reaction mechanism responsible for the peaks (FS) in the mass spectra, a pure phenomenological question arises: at what excitation energy the effect

will vanish? Our experiment was partially inspired by the fact that there were indications of a fine structure in the mass spectrum of the fragments in the reaction  $^{238}\text{U} + ^{40}\text{Ar}$  (249 MeV) (fig. 14 in ref. [11]) just at the initial excitation energy  $E^* > 70$  MeV. Our idea was to enhance these structures by selective gating on the coincident neutron spectra. In particular, pre-scission neutron emission both favours long-lived systems and provides an efficient and fast cooling mechanism necessary to bring the excitation of the composite system down to the levels where shell effects could be manifested.

## 2 Experiment

The experiment was carried out at HENDES (High Efficiency Neutron Detection System) set-up at the K-130 cyclotron at the University of Jyväskylä [12]. A 300  $\mu$ g/cm<sup>2</sup> thick <sup>238</sup>U target was prepared by evaporation of natural uranium onto a 65  $\mu$ g/cm<sup>2</sup> Al<sub>2</sub>O<sub>3</sub> backing. The target was positioned nearly perpendicular (85°)relative to the beam axis. Reaction fragments were detected by a two-arm time-of-flight (TOF) spectrometer. It consisted of two, 24 cm in diameter, position-sensitive avalanche counters (PSAC) and a single parallel plate avalanche counter (PPAC) working as a start detector. Position resolution of

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Fig. 1. Contour plot of the double-differential cross-section for the reaction  $^{238}\text{U}+^{48}\text{Ca}$  (6 A MeV) obtained in an inverse kinematics experiment [13]. With our present setup only the region between the two parallel lines is accessible.



Fig. 2. Neutron spectrum from one of the PSND located at the backward angles.

PSACs was better than 1.5 mm and the combined startstop time resolution was 620 ps (FWHM) or  $\sim 5$  amu at the 25 cm flight-path length. The PSACs were placed at  $60^{\circ}$  and  $70^{\circ}$  with respect to the beam axis covering  $90^{\circ}\pm30^{\circ}$  in the center-of-mass frame. As can be inferred from fig. 1, the yield of unequilibrated mass-drift modes is kinematically suppressed for the chosen detection geometry [13]. A single,  $20 \text{ cm}^2$  surface-barrier Si detector was located behind one of the PSAC to measure the energy of the fragments. Neutrons were detected by 5 positionsensitive neutron detectors (PSND)located approximately at 0.5 meters distance around the target position. Each PSND had 1 m active length, 1.4 ns time resolution and 10–20 cm position resolution. A typical neutron energy spectrum from one of the PSNDs on the back is shown in fig. 2. To produce this spectrum a standard pulse-shapebased  $n/\gamma$  separation was applied and TOF values were converted into energy.



Fig. 3. TKE vs. mass plotted for all registered events. Since our detectors cover only part of the full solid angle and have, because of kinematics, reduced sensitivity to high-TKE events, the mean value of TKE on the plot is 150 MeV instead of 217 MeV.

The quality of the fission fragment data can be judged from the total kinetic energy (TKE) vs. mass plot shown in fig. 3. The mean TKE value of 150 MeV as seen on the plot is significantly less than the 217 MeV predicted by the systematics [14]. This discrepancy does not result from a calibration shift or from a similar mistake since the TKE values associated with elastic scattering are correct. The difference results from the restricted angular coverage of the fission fragment detectors. Also, the nearly perpendicular position of the target further suppresses fragments with high TKE values (*i.e.* big laboratory folding angles) as they must exit almost parallel to the target's surface. Fortunately, this limitation in the detection geometry does not have any other adverse effects on our measurements.

#### 3 Results and discussion

#### 3.1 Two-body events

Figure 4a shows the integral mass spectrum for all events. It has a smooth shape, without any pronounced structures, and looks like the mass spectra obtained at similar excitations in previous works [7,8]. Figure 4b presents the same mass yields but selected only for the events fulfilling the following two conditions: 1) the folding angle is in a close proximity of  $180^{\circ}$  in the centre-of-mass frame and 2) the event is accompanied by a neutron emitted into the backward hemisphere (and detected by the backward PSND). The fine structure (FS) of the spectrum in the form of distinct peaks is clearly visible. The FS peaks are located in the vicinity of the mass numbers  $A \simeq 70-80$ , 100 and 130, which coincide with masses of spherical and deformed magic nuclei (clusters) of Ni, Ge, Sr, Zr, Mo, Sn and Te [15]. This is the first time that such a structure is observed in the decay of a composite system with initial excitation of over 70 MeV. Figure 4c shows that



Fig. 4. Fission fragment mass spectra in the reaction  $^{238}\text{U}+^{40}\text{Ar}$  ( $E_{\text{lab}} = 243$ ): (a) all fragments, (b) fragments in coincidence with neutrons emitted at backward angles, (c) fragments with low TKE values registered in coincidence with neutrons emitted at backward angles.

this FS survives up to very low TKE values with respect to the known mean TKE value. At first it seems difficult to connect FS observed in the mass yields in figs. 4b and 4c with one of the many possible reaction channels such as: quasi-fission, fast-fission, or fusion-fission. However, strong enhancement in coincidence with backward emission of neutrons is a decisive argument in favour of the slow, compound-like fission process. In our case neutrons detected in the PSND located at the backward angles are emitted predominantly from the composite system (prescission neutrons) because the post-scission component is sufficiently suppressed due to geometry, even if the postscission neutron multiplicity  $\nu_{\text{post}}$  is not negligible. As is well known, pre-scission neutron multiplicity  $\nu_{\rm pre}$  is a sensitive instrument for testing the time scale of fission [11]. Only slow processes can give rise to high values of prescission emission. Furthermore, the same is true for the deformation. Fast processes require compact shapes whereas our TKE values for the spectra with FS indicate the presence of the essential pre-scission elongation. This trend is illustrated by fig. 5 in which a clear change in the TKE yield can be observed comparing the spectrum obtained for all events (no selection) with the spectrum in coincidence with backward-emitted neutrons. The gross spec-



Fig. 5. TKE yields: (a) for all binary events, (b) for the fragments detected in coincidence with neutrons emitted at backward angles.

trum of fig. 5a corresponds to the mass spectrum 4a and the neutron-gated spectrum, to the mass spectrum 4b.

Therefore, we are quite confident that the events selected in fig. 1b are linked predominantly with the decay of a long-lived superheavy composite system  $^{278}110$ . The fact that FS does not disappear at very low TKE values further confirms the above conclusion.

Manifestation of shell effects in fission at the initial excitation of over 70 MeV would be very surprising. However, fission does not necessarily occur at the initial excitation. As it was shown for the reaction  $^{238}\text{U}+^{40}\text{Ar}$  (249 MeV),  $\nu_{\rm pre}=3.5$  [11].

Since, on average, nearly four neutrons are emitted prior to scission, it is clear that the compound system should be cold enough to manifest shell effects. We intend to perform a more detailed analysis of the reaction mechanism as soon as more data will be collected from the forthcoming experiments.

#### 3.2 Three-body events

Let us discuss a possible pre-scission shape of the compound system responsible for the peaks in fig. 1b. We could imagine it as the light magic fragment (Ni, for instance) attached to the heavy complementary fragment of elongated shape in order to ensure the observed, low TKE values. In the decay such events should well conform the binary kinematics. Alternatively, as we have already suggested in our recent study of ternary spontaneous fission [16], there could be a pair of light, magic nuclei resembling a barbell, with a massive, elongated neck connecting two spherical parts. If such a system (sketched in the upper left corner of fig. 6) should decay via double rapture of the neck, we would expect the two fragments from the ends of the barbell to fly away with roughly identical velocities in the opposite directions along the symmetry axis and the remaining "handle" to remain nearly at rest in the CM system.

173



**Fig. 6.** Energy of FF extracted from the Si-detector *vs.* TOF-TOF energy. In addition to the main group of events (labeled 1), 3 more groups can be discerned (labeled 2-4). Their presence can be explained by assuming tripartition of the compound system leading to a heavy stationary residue.

In order to check this intriguing hypothesis, we have searched for events with approximately equal velocities of both fragments and unrealistically high  $E_{\rm TOF}$  values.  $E_{\rm TOF}$  is the energy calculated from the measured velocities assuming that the mass of the two fragments adds up to that of the compound nucleus (often corrected for pre-scission emission). Naturally, this works only for binary events so if a significant mass is missing, we get significantly too high  $E_{\text{TOF}}$ . Since for part of the events we have measured fragment energy directly with semiconductor detector, we could compare  $E_{\text{TOF}}$  and  $E_{\text{Si}}$ . Events with similar mass deficit should be located along the same lines as the dependence of energy on mass is linear. Figure 6 shows the results. Four groups of points emerge with significantly different  $E_{\text{TOF}}$  energies for the same  $E_{\text{Si}}$  values. The group labelled 1 (the most intense) is linked with conventional, binary symmetric fission events. For these events in the first approximation  $E_{\rm TOF} \simeq E_{\rm Si}$ .

Using this approximate calibration, one can estimate that points labeled 4 are consistent with the hypothesis of two Ni clusters being registered leaving a heavy  $(A \simeq 138)$ residue behind. The same calculations show that points labelled 3 (fig. 6) correspond to registration of two fragments with mass about 100 amu (probably <sup>96</sup>Sr or <sup>100</sup>Zr) and points 2 may be due to the ternary decay involving  $^{96}\mathrm{Sr}$  and  $^{128}\mathrm{Sn}$  clusters. Another way to estimate the mass of these clusters is demonstrated in fig. 7. The events presented in the plot are selected using the following gates:  $V_{\rm CM}^{(1)} \simeq V_{\rm CM}^{(2)}$  (within 20% accuracy), coincidence with a neutron, folding angle at least  $170^{\circ}$  in the centre-of-mass frame. We have used the calibration procedure proposed in [17] to estimate the mass of the fragment corresponding the bottom line 1 in fig. 7. The mass number is equal to  $72\pm3$  (Ni). The sets of points 2 and 3 may be linked with fragment masses  $\sim 96$  amu (Sr) and  $\sim 130$  amu (Sn, Te).

Comparing the plots in figs. 6 and 7, one can note that the integrity of the "Ni points" is seen more vividly in fig 7.



Fig. 7. FF energy based on the Si-detector as a function of the square of the fragment velocity in the laboratory frame. Unlike fig. 6, no mass assumption was necessary. The labels of the point sets coincide with those in fig. 6.

This fact is probably due to higher  $\nu_{\rm post}$  in the double-Nimode of the decay. It is reasonable to expect a value of  $\nu_{\rm post}$  to be proportional the mass of the neck as clusters should stay unexcited [18]. All the above verifications are fully consistent with the assumption of the new type of reaction — a collinear cluster tripartition of an excited nucleus. The results relatively close to ours have been obtained very recently [19] in the reactions  ${}^{32}{\rm S}{+}^{59}{\rm Co}$  and  ${}^{32}{\rm S}{+}^{63}{\rm Cu}$  at 5.7 A MeV. The authors point to the presence of prompt ternary break-up of the composite system under study. The decay appears to occur in a collinear configuration. In spite of the large energy dissipation, some of the events shows structure effects, *i.e.* the possible presence of clustering phenomena in the reaction (at least one fragment should be an  $\alpha$ -like nucleus as suggested in [19]).

### 4 Conclusions

For the first time a pronounced fine structure (FS) in the form of distinct peaks has been observed in the neutron gated fragment mass distribution from the decay of the <sup>278</sup>110 composite system at initial excitation above 70 MeV. The FS peaks are located in the vicinity of the mass numbers  $A \simeq 70$ –80, 100 and 130, which correspond to spherical and deformed magic nuclei (clusters) of Ni, Ge, Sr, Zr, Mo, Sn, Te. The observed FS does not disappear at very low TKE values. Both this fact and the enhancement of the FS just in coincidence predominantly with pre-scission neutrons lead to conclusion that a longlived state of the composite system gives rise to the effect observed.

Some of the detected events can be treated as an indication of ternary fission via elongated pre-scission configurations leading to the emission of two clusters along the fission axis and a heavy residue nearly at rest. Hence we are inclined to believe that the collinear cluster tripartition channel proposed by us previously [16] is indeed taking place in fission of heavy nuclei. Yu.V. Pyatkov *et al.*: Peculiarities in fragment mass distribution in the  $^{238}U + {}^{40}Ar$  (243 MeV) reaction 175

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