

The role of Fission in the search of the Super Heavy Land: fission modes in heavy and superheavy nuclei. Case study of ^{180}Hg

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Abstract. Many observations strongly support the hypothesis that nuclei may fission through several independent fission modes (multimodal fission) interpreted as different prescission shapes and fission paths in a multidimensional potential energy landscape where shell effects are dominant. Mass distributions of the fission fragments are sensitive to the potential energy landscape and appear to be single humped (symmetric) or double humped (asymmetric). In many cases a mixture of both modes is observed.

We propose here our study on ^{180}Hg . Binary fission fragments formed in the reaction $^{68}\text{Zn} + ^{112}\text{Sn} \rightarrow ^{180}\text{Hg}$ at different excitation energies around the Coulomb barrier were detected using the double-arm time-of-flight technique with the spectrometers CORSET. The experiment was performed at JYFL (Jyväskylä, Finland). We will discuss an analysis of the mass distributions in terms of fission modes predicted by a five-dimensional fission model. We have found out that the mass distributions can be well reproduced by considering a symmetric fission mode and two asymmetric modes at ($A_L \approx 80$, $A_H \approx 100$) and ($A_L \approx 70$ and $A_H \approx 110$).

1 Introduction

The study of the fission process is important not only for searching pathways to synthesize new superheavy elements and to predict their stability against fission, but also for the direct impact on the understanding of the fission recycling process in r-process of nucleosynthesis. A description of the fission process with reliable predictive power is therefore needed for low-energy fission where the fission fragments mass distributions are strongly sensitive to microscopic effects.

The systematics of Mass-Energy distributions in spontaneous and low energy fission of heavy and superheavy nuclei reveals several complex features of the collective nuclear motion and striking structural effects that are matter of intense interest [1]. The understanding of such features is particularly relevant in the mass range $256 \leq A \leq 276$ and charge range from $Z=100$ (Fm) to $Z=110$ (Ds) because this region of the nuclear chart constitutes the transitional region that connects the heavy actinides with the superheavy region around $N=184$ and $Z=114$. Understanding of shell effects in this region and their implementation in the potential energy surface (PES) in the multi-dimensional collective deformation space is therefore of

peculiar interest for the search of reactions leading to superheavy nuclei of the island of stability at $N=184$ as well as to the prediction of their structural properties [1, 2].

Mass distributions (MDs) in fission are usually predominantly single or double humped (symmetric or asymmetric fission, respectively). The dominance of asymmetric fission occurs in most of the nuclei in the actinide region beyond $A=226$ up to about ^{256}Fm , and is attributed to the strong shell structure of the nascent heavier fission fragment near the doubly magic ^{132}Sn [3–5]. However, nuclei such as ^{258}Fm and $^{259,260}\text{Md}$ exhibit complex MDs, each with a narrow and a broad symmetric component with a higher and lower TKE, respectively. This phenomenon is called bimodal fission [6, 7]. For instance, the mass distribution of ^{258}Fm is symmetric, however the associated TKE distribution can be decomposed into two Gaussian distributions. The opposite holds true for the spontaneous fission of ^{252}Cf for which the mass distribution is double humped and the TKE grossly single humped. These findings cannot be explained by the Liquid Drop Model (LDM) and it is necessary to resort to shell effects. Experimental observables characterizing various fission modes are the widths of the MD peaks, the position of these peaks in the asymmetric mass division, and the total kinetic energy (TKE) of the fission fragments.

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The understanding of such features resides in the concept of multimodal fission, namely the presence of valleys due to shell closures in the multidimensional PES for different mass asymmetries and degrees of deformation of the fragments. Distinct fission modes are associated with these valleys that are navigated throughout the decay. In several cases an admixture of modes is observed [1, 5–8]. Understanding of shell effects and their implementation in the PES is therefore of peculiar interest to gain insight on the fission process, in particular, for the search of the reactions leading to superheavy nuclei of the island of stability at $N=184$ as well as for the prediction of their survival probability against fission (see [1, 2, 9] for more details).

A description of the fission process through models with reliable predictive power is therefore needed for low-energy fission where the fission fragments MDs are strongly sensitive to the PES and microscopic effects. The most extensive calculations of the fission fragments MDs were carried out by Möller et al. [10] by using a recently developed Brownian Metropolis shape-motion treatment on 5D micro-macro PES. The larger dimensional space has highlighted that the PES changes very rapidly. In particular, in the pre-actinides region, two unexpected regions of asymmetric fission, one centered at nuclei around $N = 105$ and $Z = 78$ (namely, Hg isotopes), and the other at $N = 145$ and $Z = 90$ (namely, Th isotopes), have emerged. These regions are also well separated by an area of symmetric fission which is now considered to be another transitional region. The region around Th was partially expected since it was revealed experimentally much earlier for the nuclei $^{216,220}\text{Ra}^*$ [11, 12] and $^{220,224,226}\text{Th}^*$ [13–16].

In the pre-actinide region around Hg, predominantly symmetric mass distributions have been observed. A few relevant cases are ^{195}Au , ^{198}Hg , and $^{208,210}\text{Po}$, studied by means of charged-particle-induced reactions [17–19]. However, in a study of β -delayed fission of ^{180}Tl at ISOLDE, CERN, it was observed the occurrence of asymmetric fission in ^{180}Hg [20–22], with the most probable mass numbers in the MD at $A_L \approx 80$ and $A_H \approx 100$, where A_L and A_H are light and heavy fragment masses, respectively. This result triggered the hypothesis that the fission mechanism in ^{180}Hg is different from the one postulated in the actinide region, where strong shell effects of the nascent fragments, driven by the double magic ^{132}Sn , decide the shape of the MD. For such a case, the fission of ^{180}Hg should have shown a symmetric split in two ^{90}Zr fragments ($N = 50$ shell closure in the fragments). The mechanism occurring in the fission of ^{180}Hg should therefore be different from the one in the actinide region, since strong shell effects due to the nascent fission fragments are not observed.

Considering the scarcity of data about fission in the Hg region, these findings make the case of ^{180}Hg a benchmark study for our comprehension of fission, and ultimately also a test bench of the Möller’s fission model. Indeed Möller’s model, in its 5D implementation, predicts a symmetric mode and two asymmetric modes in the MD of ^{180}Hg . Beside the asymmetric modes located at $A_L \approx 80$ and $A_H \approx 100$ and found experimentally [20], the second asymmet-

ric mode is located at $A_L \approx 70$ and $A_H \approx 110$ at the lowest excitation energy. So far this last one has not been clearly observed. There are some indication of its existence in recent works [23–25] where the fission of several Hg isotopes was investigated.

The test of Möller’s model acquires much more relevance considering that its predictions concerning the fission barriers and ground state deformations are widely used in the literature, throughout the whole Segrè chart, and in particular, in the superheavy mass region to estimate the superheavy nuclei stability against fission. Therefore, the test of this model by using reactions between heavy ions which are more easily feasible and accessible experimentally gives additional ground to the predictions of the model in an unknown region such as the superheavy island.

In such a framework, we propose here our study on the fission of ^{180}Hg . Fission fragments formed in the reactions $^{68}\text{Zn} + ^{112}\text{Sn} \rightarrow ^{180}\text{Hg}$ at different excitation energies around the Coulomb barrier were detected using the double-arm time-of-flight technique with the spectrometers CORSET [26]. The experiment was performed at JYFL (Jyväskylä, Finland). We will go through an analysis of the mass distribution in terms of fission modes.

2 The experiment

The experiment was carried out at the Physics Department of the Jyväskylä University (Finland). A beam of ^{68}Zn at 300 and 355 MeV was produced by the K-130 cyclotron. The energy resolution of the beam was 1%. The target was prepared by sputtering ^{112}Sn ($200 \mu\text{g}/\text{cm}^2$) on a $30 \mu\text{g}/\text{cm}^2$ carbon backing. The enrichment of the target was 99.9%. The experimental setup was identical to the one used for the $^{36}\text{Ar} + ^{144}\text{Sm}$ reaction [27]. The binary reaction fragments were detected by the double-arm Time-Of-Flight (TOF) spectrometer CORSET [26]. Each arm of the spectrometer consists of a compact start detector and a position-sensitive stop detector. Both detectors are based on microchannel plates. The arms of the spectrometer were set symmetrically with respect to the beam axes, at the angle of 45° . For symmetric reaction products, this configuration corresponds to the 90° angle in the c.m. system. The position resolution of the stop detectors was 0.3° . The full width at half maximum (FWHM) time of flight resolution was about 150 ps. The mass and energy resolutions of the CORSET setup were calculated from the FWHM of the mass and energy spectra of the elastically scattered particles. The resulting mass and TKE resolutions were $\pm 2u$ and $\pm 6\text{MeV}$, respectively. The extraction of the binary reaction channels, exhibiting full momentum transfer, was based on the analysis of the kinematics diagram (see [28, 29] for details). Further details on the analysis of raw data are in the PhD work by A. Pulcini [30]. Relevant reaction parameters are shown in Table 1.

Each arm of the TOF spectrometer allows to measure the hit position on the stop detector and the time of flight of the fragment from start to stop detectors. By performing such a measurement among the two fragments in coincidence, it is possible to measure the velocity and angle

$^{68}\text{Zn} + ^{112}\text{Sn} \rightarrow ^{180}\text{Hg}$					
E_{lab}	E_{cm}	V_C	Q	E_{CN}^*	B_f
[MeV]	[MeV]	[MeV]	[MeV]	[MeV]	[MeV]
355	221	172.2	-138.7	82	9.8
300	187			48	

Table 1. Table of the most relevant parameters concerning the reaction: projectile energies (both in laboratory E_{lab} , and center of mass E_{cm} reference frames); Coulomb barrier, V_C ; fusion Q value; excitation energy of the CN, E_{CN}^* ; fission barrier of the CN, B_f

of emission in 3D from which mass and energy of each fragment can be extracted by using two-body kinematics. Corrections due to energy lost in the target and in the entrance windows of the detectors are taken into account by an iterative procedure and the use of range-energy models.

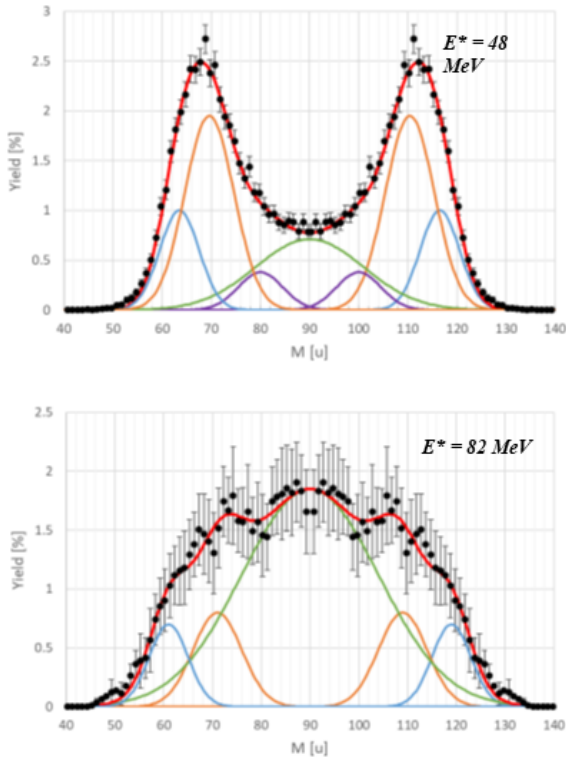


Figure 1. Mass distributions of the binary fragments from the reaction $^{68}\text{Zn} + ^{112}\text{Sn}$ at $E_{lab} = 300$ (top) and 355 MeV (bottom) as black dots with error bars. Also shown is the decomposition in symmetric (green line) and two asymmetric (orange and purple lines) fission modes. The blue line is the component associated to quasi-fission or quasi-elastic channels. The red line is the sum of all the Gaussian curves.

2.1 Mass distributions

The extraction of the mass distributions from the mass-TKE matrices is a procedure that requires several steps (see [29, 30, 32, 35] for more details). First, the velocity vectors of the two fragments in coincidence are used

to select only the events corresponding to Full Momentum Transfer (FMT). This is accomplished by using events-by-event the projections of the velocity vectors on the beam axis to calculate the center of mass velocity. For an event with FMT, this computed velocity must be around the expected center of mass velocity. The projections on the orthogonal axis is used to consider only events that are coplanar. Events that do not follow these criteria are originated by other processes (i.e. fission after transfer).

In the second step, the events with TKE smaller than the TKE of the elastic or quasi-elastic channel must be selected. Usually this is done selecting a threshold on the TKE spectrum for all the masses. In the case of the present reaction this cut-off is at values of $\text{TKE} \leq 35$ MeV.

The mass distributions obtained are shown in Figure 1 as black dots. It is evident that at both energies the mass distributions are multimodal. A combination of fission modes is possible, but also a component due to quasi-fission (QF) must be considered as the mass asymmetry of the entrance channel $\alpha_0 = (A_{target} - A_{projectile}) / (A_{target} + A_{projectile})$ is at the border of the limits where the onset of QF is expected [1]. This matter is discussed in detail in Ref. [33] where an analysis of this point is presented for the same reactions presented here by considering two reactions bringing to the same CN.

2.2 Fission modes in ^{180}Hg

To evaluate the presence of specific fission modes in the experimental data, the mass distributions calculated by Möller et al. [10] have been taken as a guideline to search for fission modes. The approach used here is the attempt to reproduce the full experimental mass distribution with the same fission modes predicted in [10], namely, the symmetric mode at $A_L = A_H = 90$ and two asymmetric modes located at $A_L \approx 80$ and $A_H \approx 100$ and at $A_L \approx 70$ and $A_H \approx 110$. This method is constrained by the fact that if a fission mode is identified, its average mass cannot change with the excitation energy being it a feature of the fissioning nucleus. If found experimentally, it is also a manifestation that such nucleus could have indeed been formed in the nuclear reaction. This is not always guaranteed being the entrance channel dynamics very important in the fusion process. For instance, the competing quasi-fission process may follow the same paths within the PES and bring to a mass split nearby the one of the fusion-fission [1].

The Gaussian curves centered on the predicted fission modes are shown in Figure 1 as solid curves. The fit is done by using the following constraints: 1) the symmetric mode has a fixed average and a width that changes according to the predictions of the Liquid Drop Model [34] (only the amplitude is a free parameter); 2) the average of each asymmetric mode is fixed by Möller's model but the widths and amplitudes are free to vary with the constraint that $A_L + A_H = 180$.

It is important to stress three points. First, the symmetric component increases its contribution with increasing excitation energy as expected by the gradual wash out of the shell effects with the increasing excitation energy. This is a known feature of the fission process. Second,

mass distributions are well reproduced considering that the fission modes do not change their mass value with the excitation energy and this is a confirmation that the same CN may have been formed at different excitation energies. Additional insight on this point can be obtained from the combined analysis of mass and TKE [8]. Third, to reproduce the experimental mass distributions at both energies an additional asymmetric component, shown as blue curves, for the most asymmetric masses, must be added. The associated masses are located around the masses of the target and the projectile nuclei, respectively, and their average masses change with the bombarding energy. This is a clear signature that this component has a direct channel origin as in the quasi-fission or quasi-elastic processes. The progress of this component with bombarding energy represents a key feature to associate that component not with a fission mode. It is a component that was not possible to eliminate with a single cut-off on the TKE. Hence, this use of the fission modes can be used to identify processes that overlap with fusion-fission events.

2.3 Conclusions

The main goal of this work is to show how a study of the nuclei in the region of masses around 180 can be of value to test models used to predict the fission and the stability of heavy and superheavy nuclei. In the physics case involving ^{180}Hg , we have measured the Mass-TKE distributions of binary reactions at two lab energies.

We have found out that the mass distribution can be well reproduced by considering a symmetric fission mode and two asymmetric modes. The asymmetric one at $A_L \approx 80$ and $A_H \approx 100$ was first experimentally found in the work by Andreyev et al. [20] by using β -delayed fission of ^{180}Tl at ISOLDE, CERN. This same mode was confirmed in Ref. [23–25, 33] by using the reaction $^{36}\text{Ar} + ^{144}\text{Sm}$ to produce ^{180}Hg and is confirmed in this work.

Furthermore, we have found out that a second asymmetric mode is revealed at $A_L \approx 70$ and $A_H \approx 110$ in agreement with the predictions of Möller's model. This result is partially in agreement with what found in Ref. [25] where three asymmetric modes are proposed, $A_L = 79, 76.5$ and 65.8 . The full agreement with this proposal is affected by at least two factors: the limited mass resolution of the TOF spectrometer and the fact that, for the reaction $^{68}\text{Zn} + ^{112}\text{Sn}$, the third mode is overlapped to the quasi-elastic component.

This is quite an interesting set of results which shows that: 1) shell effects are persistent even when the excitation energy is above ≈ 50 MeV; 2) some of the fission modes can be obscured by other reaction channels and therefore not brought to light.

There is a final important remark. The analysis of the mass distribution shown here has been performed without giving any indication of the mechanism from which those fragments are originated. By using Möller's mass distribution as a guide to constrain the search of the fission modes we are implicitly assuming that those fragments are produced by the fusion-fission channel. The search for the fission modes however requires a detailed

evaluation of the entrance channel. In this reaction it is expected a fair amount of quasi-fission, and for the case at 355 MeV, also the fast process known as fast-fission is indeed not negligible being the critical angular momentum for fusion $L_{crit} \approx 115\hbar$, bigger than the angular momentum where fission barrier is washed out $L_{B_f=0} \approx 68\hbar$. Both processes are binary reactions that produce fragments that may overlap with fusion-fission fragments. In principle quasi-fission may populate the same mass modes (we know this from the many systems studied) by traveling a path in the PES that connects the entrance channel and the exit channel without passing through the formation of the CN. Evidence of the occurrence of quasi-fission in the same reaction was indeed discussed in Ref. [33]. The contribution of fast-fission is however more difficult to estimate being this process not characterized by the presence of shell effects, as recently discussed in Ref. [35] and its occurrence is mostly indicated by the enlargement of the width of the mass distribution with respect to the predictions of the LDM.

The combined analysis of the mass modes and their strength in connection with the TKE may give us insight on this issue, namely on the amount of the relative occurrence of fusion-fission, quasi-fission and fast-fission. This aspect is crucial for a firm assignment of the fission modes and is part of a future investigation.

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References

- [1] M.G. Itkis, E. Vardaci, I.M. Itkis, G.N. Knyazheva and E.M. Kozulin, Nucl. Phys. A **944**, 204 (2015)
- [2] E. Vardaci, M.G. Itkis, I.M. Itkis, G. Knyazheva and E.M. Kozulin, J. Phys. G: Nucl. Part. Phys. **46**, 103002 (2019)
- [3] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939)
- [4] P. Möller et al., Phys. Rev. C **79**, 064304 (2009)
- [5] P. Möller et al., Phys. Rev. C **91**, 024310 (2015)
- [6] E.K. Hulet et al, Phys. Rev. C **40**, 770 (1989)
- [7] N. Carjan, F.A. Ivanyuk, Yu. Oganessian, and G. Ter-Akopian, Nucl. Phys. A **942**, 97 (2015)
- [8] T. Banerjee, E. M. Kozulin, G. N. Knyazheva, A. A. Bogachev, I. M. Itkis, E. Vardaci, A. Di Nitto, M. Ashaduzzaman, P. A. Setaro, and G. Alifano, Phys. Rev. C **108**, 064601 (2023)

- [9] K.V. Novikov et al, Phys. Rev. C **102**, 044605 (2020)
- [10] P. Möller, J. Randrup, and A. J. Sierk, Phys. Rev. C **85**, 024306 (2012)
- [11] I.V. Pokrovsky et al. Phys. Rev. C **60**, 041304 (1999)
- [12] A.Yu. Chizhov et al., Phys. Rev. C **67**, 011603 (2003)
- [13] I.V. Pokrovsky et al. Phys. Rev. C **62**, 014615 (2000)
- [14] G.G. Chubarian et al., Phys. Rev. Lett. **87**, 05270 (2001)
- [15] F. Steiper et al., Nucl. Phys. A **563**, 282 (1993)
- [16] A. A. Goverdovski et al., Phys. At. Nucl. **58**, 188 (1995)
- [17] M. G. Itkis, V. N. Okolovich, and G. N. Smirenkin, Nucl. Phys. A **502**, 243 (1989)
- [18] M. G. Itkis et al., Sov. J. Nucl. Phys. **52**, 601 (1990)
- [19] M. G. Itkis et al., Sov. J. Nucl. Phys. **53**, 757 (1991)
- [20] A. N. Andreyev et al, Phys. Rev. Lett. **105**, 252502 (2010)
- [21] J. Elseviers et al., Phys. Rev. C **88**, 044321 (2013)
- [22] V. Liberati et al., Phys. Rev. C **88**, 044322 (2013)
- [23] A.A. Bogachev et al., Phys. Rev. C **104**, 024623 (2021)
- [24] A.A. Bogachev et al., Bull. Russ. Acad. Sci., Phys. **85**, 1080 (2021)
- [25] E.M. Kozulin et al., Phys. Rev. C **105**, 014607 (2022)
- [26] E.M. Kozulin, et al., Instrum. Exp. Tech. **51**, 44 (2008)
- [27] D. Kumar et al., Bull. Russ. Acad. Sci., Phys. **84**, 1001 (2020)
- [28] I.M. Itkis, et al., Phys. Rev. C **83**, 064613 (2011)
- [29] D.J. Hinde et al., Phys. Rev. C **53**, 1290 (1996)
- [30] A. Pulcini, "The role of fission in the search of the super heavy promised land", Ph.D. thesis, Dipartimento di Fisica, Università degli Studi di Napoli "Federico II" (2020) (unpublished)
- [31] E.M. Kozulin et al., Phys. Rev. C **86**, 044611 (2012)
- [32] E. Vardaci et al., Phys. Rev. C **101**, 064612 (2020)
- [33] E.M. Kozulin et al., Phys. Lett. B **819**, 136442 (2021)
- [34] M. G. Itkis and A.Ya. Rusanov, Phys. Part. Nucl. **29**, 160 (1998)
- [35] E.M. Kozulin et al., Phys. At. Nucl. **86**, 56 (2023)