Discrimination between roles of fissioning nucleus and asymmetry degree of freedom on the even-odd structure in fission-fragment yields

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Abstract

Based on a wide systematics of fission-fragment distributions measured at Lohengrin and at GSI, the even-odd staggering in the fission-fragment nuclear-charge yields is investigated. The general increase of the even-odd staggering with asymmetry is attributed to the absorption of the unpaired nucleons by the heavy fragment. As a consequence, the well established trend of even-odd staggering in the fission-fragment charge yields to decrease with the fissility is accredited in part to the asymmetry evolution of the charge distribution. This interpretation is strongly supported by the data measured at GSI, which cover the complete charge distribution and include precise yields at symmetry. They reveal that the even-odd effect at symmetry remains constant over a broad range of fissioning nuclei.

Key words: fission-fragment yields, even-odd staggering, pairing interaction, dissipated energy

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1. Introduction

Even-odd staggering is a well-known characteristic of low-energy fission-fragment yields. The large amplitude of this staggering, which may reach 40% in the case of thorium [1], has always been fascinating to nuclear physicists. The observation of odd-Z fragments from an even-Z fissioning nucleus testifies the

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re-organisation of the intrinsic structure of the fissioning nucleus on its path towards scission. Indeed, in thermal-neutron induced fission of even-Z actinides, the compound nucleus reaches the saddle deformation with an intrinsic excitation energy below the pairing gap. Thus, an ensemble of fully paired protons undergoes the deformation down to scission, where at this stage at least one pair is broken and both unpaired protons end up in different fission fragments to produce odd-Z species. The amplitude of the even-odd staggering is therefore linked to pair breaking and dissipation during the deformation in the fission process. It has always been a challenge to understand the mechanism of dissipation and the relation between pairing effects and the dissipated energy. The present work focuses on data for which nuclear charge yields are accessible. They emerge from experiments performed at Lohengrin for fissile actinides and at GSI for a long chain of neutron-deficient Th isotopes. This ensemble of data allows accessing an unprecedented systematic on pairing effects in fission-fragment yields.

2. Global even-odd staggering and dissipated energy

A simple method to quantify the pairing effect in the fragment production is to define the global even-odd staggering $\delta$ as the average difference between even- and odd-Z yields over the full available distribution [1]. Several models have attempted to quantitatively relate the global even-odd staggering to the dissipated energy. The model proposed by Nifenecker et al. [2] is certainly the most widely used as it correlates $\delta$ and the dissipated energy $E_{\text{diss}}$ with a very simple expression. This model considers a maximum number of broken pairs at scission, which is determined as the ratio between the dissipated energy and the amount of energy necessary to break a pair (the pairing gap). The nucleus at scission is then considered as a fully paired core and an ensemble of broken pairs, to which a combinatorial analysis is applied in order to determine the probability to break a proton versus a neutron pair, the probability to break a pair if the necessary amount of energy is gained, and finally the probability that the nucleons of the broken pair end up in two different fragments. This model estimates a dissipated energy of about 4 MeV for $^{230}\text{Th}$ to 12 MeV for $^{250}\text{Cf}$.

An alternative approach by Bouzid et al. [3] is based on dynamical considerations of the fission process. In this model, the descent from saddle to scission is considered to be adiabatic, and the violent neck rupture leading to the formation of the two separated fragments causes the pair breaking. The probability to break a pair is correlated with the velocity of the neck rupture, which is shown to increase with the Coulomb repulsion at the scission point. The Coulomb repulsion
is linked to the Coulomb parameter $Z_c = Z_f^2/A_f^{1/3}$, where $Z_f$ and $A_f$ are the nuclear charge and mass of the fissioning nucleus. The probability that the broken pairs end up in different fragments is a parameter fitted to reproduce the data. In particular, this model considers the difference in the proton- and neutron-number staggering, as a consequence of the less violent neck rupture for protons, since they are less present in the neck due to the Coulomb repulsion.

A rigorous formulation for the even-odd staggering has been derived by Rejmund et al. [4] in the frame of the statistical model. It is based on a realistic description of the number of quasi-particle excitations of the proton and neutron sub-systems as a function of the excitation energy at scission. The pairing-gap parameter depends on excitation energy, deformation, and the number of quasi-particles. In the model of Nifenecker et al., the number of broken pairs is deduced from energy consideration, convoluted to a fitted parameter. In the model of Rejmund et al., the number of broken pairs is based on a rigorous description of the available single-particle levels.

In the measurement of fragment yields of fissile actinides at Lohengrin, it has been observed that the global even-odd staggering decreases with the fissility of the fissioning system [5]. The amplitude of the even-odd effect being associated to the dissipated energy gained by the nucleus, the decrease of the even-odd staggering with fissility has brought up the idea that more energy is dissipated in the descent from saddle to scission as the fissility of the fissioning nucleus increases. The fissility parameter $x = Z_f^2/A_f$ reflects the stability against fission, but its connection with the dissipated energy is not clear. On the other hand, the total energy release from saddle to scission is proportional to the Coulomb parameter $Z_c$ [1, 6] and therefore, the evolution of the even-odd staggering with the fissioning nucleus is usually investigated as a function of the Coulomb parameter [4, 7]. The evolution of the global even-odd staggering is displayed as a function of the fissility or Coulomb parameters in Fig. 1, left and right panels, respectively. A clear decrease is observed as a function of both parameters.

### 3. Local even-odd staggering and influence of the asymmetry

The local even-odd staggering $\delta(Z)$ is a measure of the deviation of the nuclear charge distribution from a smooth behaviour, and is usually studied following the prescription of Tracy et al. [8]. For different fissioning systems [1, 9, 10, 11], the even-odd staggering has been shown to be larger for large asymmetry. This experimental observation, reproduced in Fig. 2, has led to the notion of cold asymmetric fission, for which extreme deformation would take most of the
available excitation energy, and consequently the intrinsic excitation energy would remain low \([9]\). However, no elaborate model exists to quantitatively describe the even-odd effect based on these assumptions.

The general understanding of the even-odd effect briefly depicted above has been perturbed by the discovery in the late 90’ s of a large even-odd effect in the \(Z\) distributions of odd-\(Z\) fissioning nuclei \([12]\). In these odd-\(Z\) systems, the probability to have at least one unpaired proton is always one. Assuming that any unpaired proton ends up in one or the other fragment with equal probability, it was expected that the even-odd effect would be zero over the full \(Z\) distribution. With experimental techniques based on inverse kinematics, the even-odd staggering has been measured for a large systematics of actinium and protactinium isotopes, over the complete \(Z\) distribution. Its value was found to be zero close to symmetry and systematically increasing for large asymmetry, up to amplitudes as large as 40\%. In the heavier part of the fragment distribution, the even-odd staggering was found to be negative, revealing a higher probability for the unpaired nucleon to end up in the heavy fragment. The same observation of large even-odd effect has been reported in neutron-induced fission of \(^{237}\)Np \([13]\), and discussed in term of energy balance; as the pairing gap is decreasing with the fragment mass, the light fragments remain paired preferably. However, only the binding energy of a cold system is considered, without any intrinsic excitation. A statistical description of the even-odd staggering with the asymmetry based on the level density of the fission fragments formed at scission \([12]\) reproduces the larger probability for the unpaired nucleons to end up in the heavy fragment. This model gives a quantitative prediction of the general increase of the even-odd staggering with the asymmetry, for odd-\(Z\) fissioning nuclei as well as for even-\(Z\) fissioning nuclei.

The observation of an even-odd staggering for odd-\(Z\) fissioning nuclei and its interpretation reveal that the relation between the amplitude of the even-odd staggering in fission-fragment charge yields and the intrinsic excitation energy at scission is not as direct as suggested by the models discussed before. Indeed, neither the statistical description of Nifenecker et al. nor the dynamical description of Bouzid et al. can explain the appearance of an even-odd structure for odd-\(Z\) fissioning nuclei since in these models, the probability of unpaired nucleons to end up in one or the other fragment does not depend on the size of the fragments. Only the statistical model of Rejmund et al. considers both the effect of dissipated energy in symmetric scission, and the influence of asymmetry in the fission-fragment yields explicitly.
4. Evolution of the fragment distribution with the fissioning nucleus and its influence on the even-odd staggering

In low-energy fission of most actinides, shell effects induce an asymmetry in the mass and charge distributions of fragments, which show two groups [14]. The group of heavy fragments is distributed over an average value of $A \sim 140$, independently of the mass of the fissioning system. In order to keep the total mass conservation, for heavier fissioning nucleus, the group of the light fragments moves towards heavier masses, approaching gradually the symmetry. The equivalent behaviour is observed in charge distributions. Precise information has been obtained for the light part of the asymmetric fission-fragment distribution at the Lohengrin spectrometer [1, 9, 10, 11, 15, 16, 17, 18, 19, 20]. They are reported in Fig. 2 left. They show an average light charge centred on values varying from $Z = 36$ to 44, when considering fissioning nuclei from Th to Cf. This coincides with a heavy charge distribution centred on a constant value $Z = 54$, independently of the fissioning nucleus, as was already reported in [21]. In the right part of Fig. 2 the associated local even-odd staggering is displayed, revealing a general trend to decrease towards symmetry. Even though $^{240}\text{Pu}$ and $^{234}\text{U}$ show a rather constant behaviour, even-odd staggering as large as 50% is observed for large asymmetry in $^{246}\text{Cm}$, $^{236}\text{U}$, and $^{230}\text{Th}$. The unavoidable correlation between fissility and symmetry raises the question whether the previous picture of decreasing even-odd structure with fissility or Coulomb parameter is an effect of increasing dissipation or of increasing symmetry in the distribution. To answer this question, it is necessary to investigate the evolution of the local even-odd effect with the fissioning system for symmetric and asymmetric splits independently.

To explore the fission asymmetry, it is convenient to define the asymmetry parameter: $a = (Z_H - Z_L)/Z_f$, where $Z_H$ and $Z_L$ are the charge of heavy and light fragment, respectively. In Fig. 3 the local even-odd staggering in $Z$ distribution of fissile actinides is displayed as a function of the fissility and the Coulomb parameter of the fissioning nucleus, for different values of the asymmetry (open squares) and compared to the values close to symmetry (full squares). These data cannot access the symmetry $a = 0$, as seen in Fig. 2. Therefore, the investigation of the evolution of the local even-odd staggering at symmetry is limited to the evolution at the most reachable symmetry, which progressively approaches the symmetry as the fissility or the Coulomb parameter increases. Actually, for these most symmetric points, the asymmetry parameter ranges from $a = 0.14$ for Th to $a = 0.05$ for Cf. Consequently, the fissioning nucleus and the asymmetry parameters are correlated, and no conclusion can be drawn on their respective influences on the
evolution of the even-odd staggering close to symmetry. This shows the importance of the new experimental technique based on inverse kinematics [22], which gives access to the complete Z distribution, and thus provides precise data at symmetry. These data sets are included in Fig. 3 for different values of the asymmetry (open circles) and at symmetry (full circles). For large asymmetry (upper panels of Fig. 3), although fission is induced with moderately higher excitation energy in the inverse kinematics technique, GSI and Lohengrin data coincide and show the same trend. The amplitude of the even-odd staggering is decreasing with fissility or Coulomb parameter. However, for symmetric splits, the local even-odd staggering measured in GSI shows a remarkable constant amplitude. For fissile nuclei, the even-odd staggering close to symmetry shows a moderate decrease, which is probably the result of the correlation between the most symmetric point experimentally observable, and the fissility (or the Coulomb parameter) of the fissioning nucleus. When the asymmetry is lowered (shown in the middle and lower panels of Fig. 3), the slope of the local even-odd effect reduces and gradually approaches the constant value of the even-odd staggering at symmetry. It is the first time that the local even-odd staggering at symmetry is reported to be essentially the same for the whole range of fissioning nuclei considered. The variation of the global even-odd effect seems to be caused to a great extent by the higher probability for the unpaired protons to end up in the heavy fragment.

In the right panels of Fig. 3, the local even-odd staggering is displayed for different values of asymmetry as a function of the Coulomb parameter. At symmetry, the same constant trend is observed. However, in this case, the large systematics measured in GSI is restricted to a narrow range of the Coulomb parameter compared to the fissile nuclei studied at Lohengrin, which show a moderate decrease. As discussed above, the Coulomb parameter is understood to be more adequate to relate to the total energy release from saddle to scission, and thus to the dissipated energy. Therefore, the constant behaviour observed when displayed as a function of the fissility may be reviewed as an artefact produced by the choice of a wrong ordering parameter. On the other hand, the data for fissile isotopes equally suggest the constancy of the even-odd staggering at symmetry. Indeed, as pointed out above, the most symmetric data available for fissile isotopes are correlated to the Coulomb parameter. When the Coulomb parameter increases from 1322 to 1525, the most symmetric data correspond to asymmetry progressively decreasing from $a = 0.14$ to $a = 0.05$. As demonstrated in Fig. 2, the even-odd staggering is decreasing towards symmetry, and therefore the even-odd effect expected at symmetry for these nuclei is systematically lower than the one reported. In addition, the decrease is expected to be larger for small Coulomb parameter (larger asym-
metry) than for large Coulomb parameter (smaller asymmetry). Consequently, the moderate decrease observed for fissile isotopes cannot be taken for the trend at symmetry, which is expected to be very close to a constant value.

As discussed in references [4, 12], the even-odd effect at symmetry is not influenced by a higher probability of unpaired protons to end up in one of the fragments. At symmetry, the production of odd-Z fragments from an even-Z fissioning nucleus reveals the probability that at least one pair has been broken in the descent from saddle to scission. Consequently, the even-odd staggering is directly connected with the amount of dissipated energy. The constant value of the even-odd effect at symmetry has thus strong implications on the understanding of dissipation in fission, as it suggests that the dissipated energy is essentially independent of the fissioning nucleus, in contradiction to the previous understanding.

5. Conclusion

A systematic investigation on even-odd staggering in fission-fragment yields based on thermal-neutron-induced fission of fissile nuclei and electromagnetic-induced fission of neutron-deficient thorium isotopes is reported. It is shown that the global even-odd effect cannot be considered to derive conclusions on dissipation, as it includes a strong contribution of the asymmetry of the scission split, which has to be taken into account to explain the amplitude of the even-odd effect. As a consequence, the previous models [2, 3], which aimed at describing the even-odd structure without considering the influence of the fission-fragment phase space, are not suitable to derive realistic conclusion on the dissipated energy. Finally, the local even-odd effect at asymmetry measured in thermal-neutron induced fission or in electromagnetic-induced fission show a similar trend to decrease with increasing fissility or Coulomb parameter of the fissioning nucleus. The local even-odd effect at symmetry is shown to be essentially independent of the fissioning system, which forces to revisit the previously established influence of the fissioning system on dissipation.

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Figure 1: Global even-odd effect as a function of the fissility parameter (left) or the Coulomb parameter (right). Data are from thermal-neutron induced fission of $^{249}$Cf $^{19, 10}$, $^{245}$Cm $^{15, 11}$, $^{239}$Pu $^{18}$, $^{235}$U $^{16, 9}$, $^{233}$U $^{17}$ and $^{229}$Th $^{20}$.

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Figure 2: Left: Z distribution for thermal-neutron induced fission of \(^{230}\text{Th},^{236}\text{U},^{234}\text{U},^{240}\text{Pu},^{246}\text{Cm}\) and \(^{250}\text{Cf}\), from bottom to top. A dashed line indicates symmetric split. Right: the corresponding local even-odd effect shown as a function of Z. The fissility x and the Coulomb parameter \(Z_c\) of the fissioning nucleus are indicated.
Figure 3: Upper panels: Local even-odd effect as a function of the fissility $x$ (left) and Coulomb parameter $Z_c$ (right), for an asymmetry of 0.22 (open symbols), and compared to the local even-odd effect measured at symmetry (full symbols). Data measured in inverse kinematics on neutron-deficient Th isotopes are shown as circles, data on fissile isotopes measured at Lohengrin as squares. Middle and bottom panels: same as above with asymmetry of 0.18 and 0.14, respectively. Lines are to guide the eye.