Revisiting the even-odd staggering in fission fragment yields

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REVISITING THE EVEN-ODD STAGGERING IN FISSION-FRAGMENT YIELDS

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Abstract. The even-odd staggering observed in the experimental fission-fragment nuclear-charge yields is investigated over a wide systematics of fission fragments measured at Lohengrin in direct kinematics and at GSI in inverse kinematics. The general increase of the even-odd staggering in the fission-fragment charge yields towards asymmetric charge splits is explained by 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 attributed 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 around symmetry remains constant over a large range of fissility.

INTRODUCTION

Even-odd staggering in fission fragment yields has been observed since the first experiments investigating fission fragment properties at low excitation energies. The large amplitude of this staggering, which may reach 40% in the case of thorium [1], has always been fascinating to nuclear physicists. The reason of this fascination is that it is connected to fundamental properties of nuclei, as pairing interaction between nucleons and dissipation in the deformation process. The observation of odd-Z fragments from an even-Z fissioning nucleus testifies the re-organisation of the intrinsic structure of the fissioning nucleus on its path towards scission. Indeed, in the 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 nucleons end up in different fission fragments to produce odd-Z fragments. The even-odd effect is therefore an experimental evidence for the pair breaking and dissipation during the deformation. It has been always a challenge to understand the mechanism of dissipation and the relation between the even-odd effect and the dissipated energy. The even-odd effect has been investigated widely, and mainly in experiments where the charge distribution may be measured. Indeed, mass distributions, even though showing a reminiscence of the strong even-odd effect in proton number,

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are less easy to interpret, as they are the combined result of the even-odd effect in proton and in neutron numbers, which do not have the same amplitude, and in addition the even-odd staggering in neutron number is being influenced by the neutron evaporation of highly deformed fragments formed at scission. In the present contribution, we concentrate on data for which nuclear charge yields are accessible. They emerge mainly 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 systematics on the pairing effect in fission-fragment yields.

Influence of the fissility of the system

In the measurement on fissile actinides in Lohengrin, it has been observed that the even-odd staggering decreases with the fissility of the fissioning system [1]. As mentioned above, 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. Several models have attempted to quantitatively relate the even-odd staggering with the dissipated energy. The most widely used model is certainly from Nifenecker et al. [2], as it gives a very simple expression of the even-odd staggering as a function of the dissipated energy. 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 combinatory 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 (it is possible to excite a pair as well), and finally the probability that the nucleons of the broken pair end up into two different fragments. In conclusion, the simple expression between the even-odd staggering and the dissipated energy is:

$$E_{diss} = -4\ln(\delta_Z) \qquad (1)$$

which corresponds to a dissipated energy of about 4 MeV for ^{230}Th to 12 MeV for ^{250}Cf. A more accurate formulation in the frame of the statistical model for the even-odd effects as a function of the excitation energy has been derived by Rejmund et al. [3]. 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 standard 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.

A different, and widely acknowledged, approach by Bouzid et al. [4] is based on dynamical considerations of the fission process. In this approach, 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. In this model, the probability to break a pair is linked to the velocity of the neck rupture, which is shown to increase with the fissility of the system, and 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.

Influence of the fission-fragment distribution

The local even-odd staggering is a measure of the deviation of the fragment distribution from a smooth behaviour as a function of Z, and is usually studied following the prescriptions of Tracy et al. [5]. It has been shown that the even-odd effect is larger for large asymmetry for different fissioning systems [1, 6, 7, 8]. This experimental observation 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 very low. In the dynamical description of fission process, the neck rupture of very asymmetric configurations is expected to be slower, and therefore less dissipative, than in symmetric configurations. However, no elaborate model exists to describe quantitatively the even-odd effect based on these assumptions.

EVEN-ODD STAGGERING IN FISSION FRAGMENT YIELDS OF ODD-Z FISSIONING NUCLEI

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 distribution of odd-Z fissioning nuclei [9]. In these odd-Z systems, the probability to have at least one unpaired proton is always one. Assuming, as usually done, that the unpaired protons will end 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 effect was found to be negative, revealing a higher probability for the unpaired nucleon to end up in the heavy fragment. This may be explained by the different number of states available for the unpaired nucleons in the fission fragments, which had not been taken into account in the previous interpretations discussed in the introduction of the present contribution. The same observation of large even-odd effect has been reported in neutron-induced fission of ^{237}Np [10]. A statistical description of the even-odd staggering with the asymmetry based on the level density of the fission fragments formed at scission [9] 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 magnitude of the even-odd staggering in the fission-fragment element yields and the intrinsic excitation energy at scission is not so direct as suggested by the models discussed before. Indeed, neither the statistical description of Nifenecker et al. nor the the dynamical description of Bouzid et al. can explain the appearance of an even-odd structure for odd-Z fissioning nuclei since the probability of unpaired nucleons to end up in one or the other fragment does not depend on the size of the fragment.

EVOLUTION OF THE Z DISTRIBUTION WITH FISSILITY AND ITS CONSEQUENCE ON THE EVEN-ODD STAGGERING

It is well known from low-energy fission of most actinides that the mass and charge distributions of fission fragments are asymmetric, showing two groups. The group of heavy fragments is distributed over an average value of A~140, independently of the mass of the fissioning system. This well-known characteristic is attributed to shell effects and the associated asymmetric modes in fission [11]. The light fragment group is centred on a shifting value as the mass of the fissioning system varies, in order to compensate for the total mass conservation. When the mass (or the fissility) of the fissioning nucleus increases, the group of the light fragments moves towards heavier masses, approaching gradually the symmetry. In the same manner, the equivalent behaviour is observed in Z distributions. However, experimental information on the heavy Z distribution is very scarce, due to the technical difficulties that arise from the detection of heavy fission fragments. Precise information has been obtained at the Lohengrin spectrometer, where isotopic and therefore Z distributions could be measured for the light part of the asymmetric fission-fragment distribution [1, 7, 8, 12, 13, 14, 15, 16, 17]. They are reported in Figure 1, left. They show an average light charge that correspond to a charge distribution centred on Z=54 for the heavy fragments, independently of the fissioning nucleus. Therefore, when considering fissioning nuclei from Th to Cf, the Z distribution of the light fragments is centred on values varying from Z = 36 to 44. In Figure 1, for each Z distribution the symmetric split is indicated with a dashed-line. In the right part of Figure 1, the local even-odd effect is shown for the different systems considered. Even though ^{240}Pu and ^{234}U show a rather constant behaviour, even-odd effects as large as 50% are observed for large asymmetric splits in ^{246}Cm, ^{236}U, and ^{230}Th. The local even-odd effect shows a general trend to decrease towards symmetry. The unavoidable correlation between fissility and symmetry raises the question whether the previous picture of decreasing even-odd structure with fissility is an effect of increasing dissipation or of increasing symmetry in the distribution. To answer this question, it is necessary to separate the parameters of asymmetry and fissility by examining the evolution of the local even-odd effect for symmetric and asymmetric splits independently with fissility.

This is done in Figure 2, where the even-odd staggering is plotted as a function of the fissility of the fissioning nucleus. The global even-odd effect shows the known trend to decrease with increasing fissility. The local even-odd effect at the value of the most
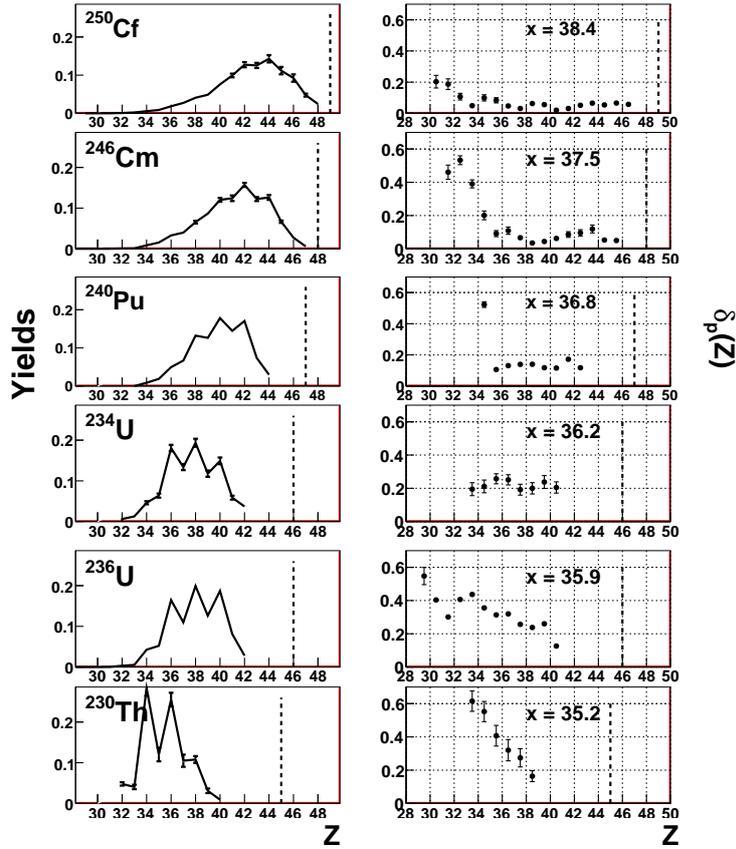
FIGURE 1. Left: Z distribution for thermal-neutron induced fission of ^{230}Th, ^{236}U, ^{234}U, ^{240}Pu, ^{246}Cm and ^{250}Cf, from bottom to top. Right: the corresponding local even-odd effect shown as a function of Z. The fissility parameter of the fissioning nucleus is indicated. In both sides a dashed line indicate symmetric split.

probable charge (ranging from 36 to 44) shows a similar trend to decrease with fissility. This is not a surprise since the local even-odd effect measured at the most probable asymmetry contributes the most to the global even-odd effect. As seen in Figure 1, the symmetry is not reachable in these experiments; therefore the empty circles in Figure 2 correspond to the *most reachable* symmetry, which in fact progressively approaches the symmetry as the fissility of the nucleus increases. For these most symmetric points, the asymmetry parameter $a = (1 - \frac{Z}{Z_{CN}})$, where Z_{CN} is the nuclear charge of the fissioning nucleus, and Z of the fission fragment, ranges from $a = 0.57$ for Th to $a = 0.53$ for Cf (while a symmetric split correspond to $a = 0.5$). Consequently, for the evolution of the even-odd effect close to symmetry in Figure 2, the fissility and the asymmetry parameters are still convoluted, and no conclusion can be drawn on the respective influences of the fissility and the asymmetry. This shows again the importance of the new experimental techniques based on the inverse kinematics [18], in which the complete Z distribution could be measured for the first time.
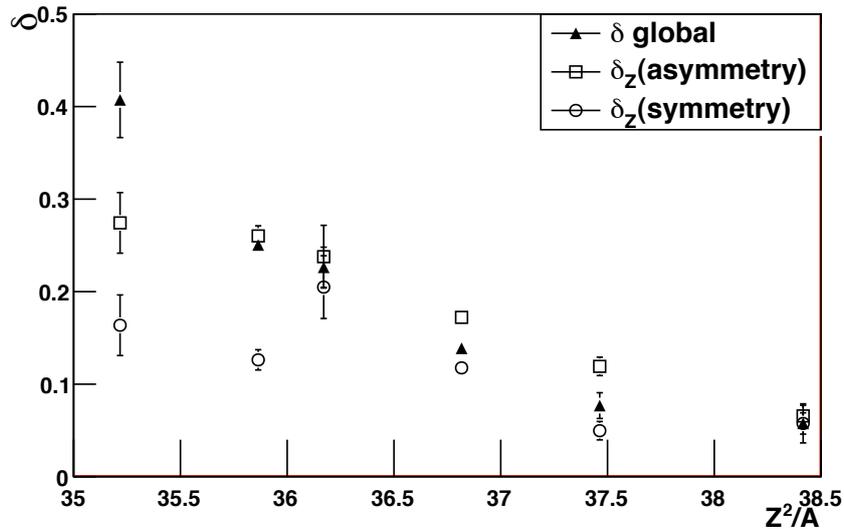
FIGURE 2. Triangles: Global even -odd effect as a function of the fissility of the fissioning nucleus. Open squares: local even-odd effect at the most probable charge split. Open circles: Local even-odd effect at the most reachable symmetry.

These data sets are shown in Figure 3, where the local even-odd effect is displayed as a function of the fissility of the fissioning nucleus, for different values of the asymmetry (open circles) and always compared to the values at symmetry (full circles). The data of Lohengrin are included (open squares), and the symmetry correspond in this case to the *most reachable* symmetry, as in Figure 2 (full squares). Figure 3 shows that the data of GSI and of Lohengrin coincide and show the same trend. For large asymmetry (upper panel of Fig. 3), the amplitude of the even-odd staggering is decreasing with the fissility. However, for symmetric splits, the local even-odd effect shows a remarkable constant behaviour. When the asymmetry is lowered towards symmetry (shown in the middle and in the lower panels of Fig. 3), the slope of the local even-odd effect as a function of fissility reduces and gradually approaches the constant value of the even-odd effect at symmetry. As shown in reference [3, 9], the even-odd effect at symmetry, where both fragments have the same propensity to absorb unpaired nucleons, is reflecting the probability that no proton pair has been broken, and therefore is directly connected to the dissipated energy from saddle to scission. The constant behaviour of the even-odd effect at symmetry has conclusively strong implications on the understanding of dissipation in fission, as it suggests that the dissipated energy is independent of the fissility, in contradiction to the previous understanding. The evolution of the even-odd staggering with fissility and asymmetry is not yet understood completely, but it is clear from the present investigation that these two parameters have to be decoupled, and that the effect of the phase space in the fission fragments has to be taken into account in order to deduce the dissipated energy.
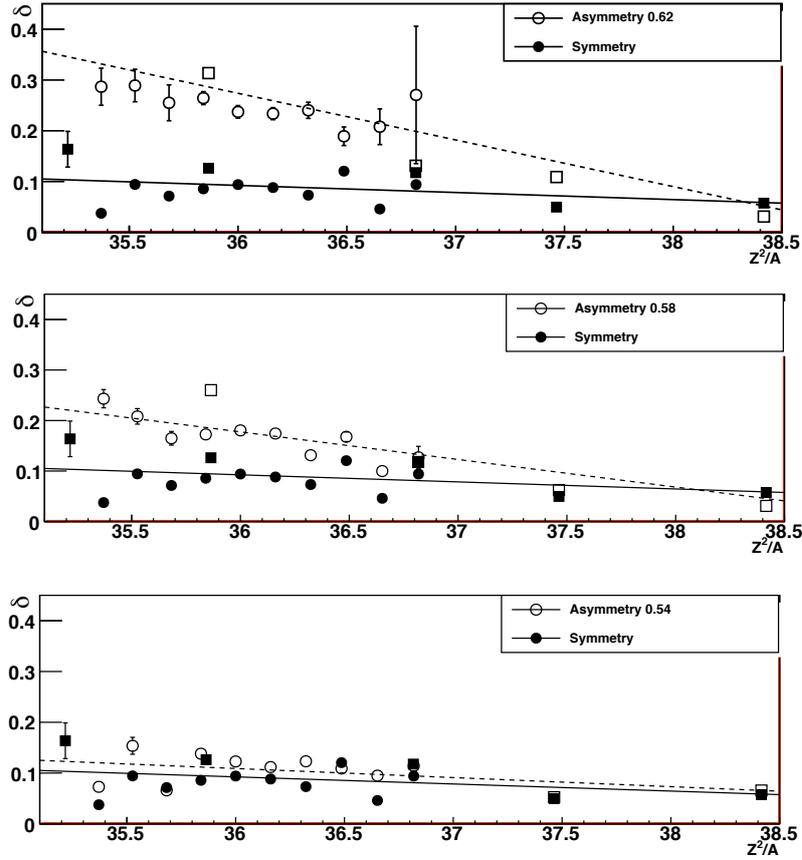
FIGURE 3. Up: Local even-odd effect as a function of the fissility parameter, for an asymmetry of 0.62 (open symbols), and compared to the local even-odd effect measured at symmetry (full symbols). Middle and bottom: same as above with an asymmetry of 0.58 and 0.54 respectively. Lines are to guide the eye.

CONCLUSION

A phenomenological investigation on even-odd staggering in fission-fragment yields based on thermal-neutron induced fission of transuranium nuclei and electromagnetic-induced fission data of neutron-deficient thorium isotopes has been presented. 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, 4], which aimed at describing the even-odd structure without considering the influence of the fission fragment phase space, become obsolete and they are shown to derive no satisfactory conclusion on the dissipated energy. Finally, the data obtained in direct kinematics (thermal-neutron induced fission) or in inverse kinematics (electromagnetic-induced fission) coincide over the full range of fissility parameter. Both data sets show a similar trend of the local even-odd effect at asymmetry to decrease with increasing fissility of the fissioning nucleus. The local even-

odd effect at symmetry, however, is shown to be independent of the fissioning system, which forces to revisit the previously established relationship between dissipation and fissility.

ACKNOWLEDGMENTS

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