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Even-odd effect in fission-fragment Z yields - a new kind of nuclear clock

Karl-Heinz Schmidt[1], Beatriz Jurado

CENBG, CNRS/IN2P3, Chemin du Solarium B.P. 120, 33175 Gradignan, France

Abstract

It is shown that the recently proposed sorting of intrinsic excitation energy in fission dynamics under the influence of pairing correlations explains the complex features of even-odd structure in fission-fragment yields. A schematic dynamical scenario is proposed. It reveals that the variation of the even-odd effect with the Coulomb parameter of the compound nucleus and the increase towards asymmetric splits carries information on several dynamical times. This new insight stresses the importance of nuclear fission as a laboratory for studying the dynamics of non-equilibrium processes in mesoscopic objects under the influence of residual interactions.

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1. Introduction

Long-range correlations in matter give rise to complex ordering phenomena. Superconductivity in solids and superfluidity in liquids are the most prominent examples of these phenomena in macroscopic systems. These ordering patterns lead to drastic changes of some bulk properties, e.g. vanishing electric resistance, respectively zero viscosity. Also in mesoscopic systems, some kind of long-range ordering exists. Pairing correlations in atomic nuclei are a prominent and well known example. They manifest themselves in an increased binding energy of nuclei with an even number of protons and/or neutrons, in a pairing gap, that

[1]* Corresponding author.
E-mail address: schmidt-erzhausen@t-online.de.

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means an exceptionally high energy of the first excited single-particle state in nuclei with even numbers of neutrons and protons, in a low momentum of inertia of nuclei at low excitation energy and angular momentum, and many other observables. However, even to date there is no consistent theory of pairing correlations in nuclei available, and the traditional methods of solving the nuclear pairing problem are borrowed from macroscopic physics (Zelevinsky, 2005)). The need for better understanding the microscopic origin of nuclear pairing (Baldo et al., 2010) explains the high interest in gathering new experimental information on the signatures of pairing correlations in nuclei.

In contrast to macroscopic systems, mesoscopic systems offer another very interesting possibility for investigating the properties of the long-range ordering pattern: Since they form a leptodermous object of well defined shape and their size is comparable with the pairing coherence length, the interplay of long-range ordering and shape changes carries very interesting information. One may even study the influence of long-range correlations on the interaction between different objects when they are brought into contact or on the dynamics of the process when one object is divided in two. These dynamical aspects of pairing correlations in nuclei have intensively been studied by the transfer of nucleon pairs in peripheral nuclear collisions at low energy (von Oertzen and Vitturi, 2001; Szilner, 2005; Peter et al., 2003) and by the appearance of a rich variety of even-odd structure in nuclear fission (Nifenecker et al., 1982; Gönnenwein, 1991; Steinhäuser et al., 1998; Rejmund et al., 2000). Experiments on nuclear transfer (Szilner, 2005; Peter et al., 2003) have indeed provided evidence for enhanced neutron-pair transfer, interpreted as a "super-current" just as in the case of a super-current carried by "Cooper pairs" (electrons) in superconductive solids. These results have been the most direct signatures of dynamical effects caused by pairing correlations in nuclei.

Nuclear fission provides a number of observables, which are modulated by an even-odd structure (Gönnenwein, 1991). Some of those will be discussed below in more detail. In most cases, the models which were proposed to interpret the complex observations remained on the level of statistical considerations up to now. Only few attempts have been made to establish a link to the dynamical evolution of the fissioning system. In this contribution, we give a new interpretation of the manifestations of pairing correlations in fission which remained unexplained since several decades. Our interpretation is directly based on several dynamical processes during the evolution of the fissioning system towards scission. This way, the even-odd effect in fission is established as a new kind of nuclear clock.

2. Experimental systematics

The most prominent manifestation of pairing correlations in nuclear fission is the enhanced production of even-Z elements in low-energy fission of an even-Z compound nucleus. Figure 1 shows the Z distribution observed in the fission of ^{229}Th, which was produced as a secondary beam from 1 A GeV ^{238}U projectiles and which was excited in the Coulomb field of lead target atoms slightly above the fission barrier with a width of about 5 MeV (FWHM) (Schmidt et al., 2000). Due to the inverse kinematics, an excellent Z resolution has been achieved. Moreover, this experiment allowed measuring the even-odd structure continuously over a large range of mass splits. This was not possible in heavier actinides due to the extremely low yields for symmetric splits.

At present, several systematic features have been established experimentally (Rejmund et al., 2010). The left part of Fig. 2 shows experimental data on the local even-odd effect (Tracy et al., 1972) as a function of charge asymmetry for different fissioning nuclei measured at ILL Grenoble. Fission was induced by thermal neutrons with the exception of ^{229}Th, where fission was induced by electromagnetic excitations. The experimental data from previous compilations (Bocquet and Brissot, 1989; Steinhäuser et al., 1998; Naik et al., 2007) and from figure 2 clearly illustrate several features:

(i) The amplitude of the even-odd structure decreases with increasing initial excitation energy and with increasing mass of the fissioning system.

(ii) There is a drastic increase of the even-odd structure at large asymmetry.

(iii) Also odd-*Z* fissioning systems like ^{239}Np and ^{244}Am show an even-odd structure in the *Z* yields, however, only at large asymmetry. Enhanced production of even-Z nuclei is observed in the light fragment, while the production of odd-*Z* nuclei is enhanced in the heavy fragment. The magnitude of the even-odd effect observed at large asymmetry is about the same in even-*Z* and in odd-*Z* fissioning systems of comparable mass.

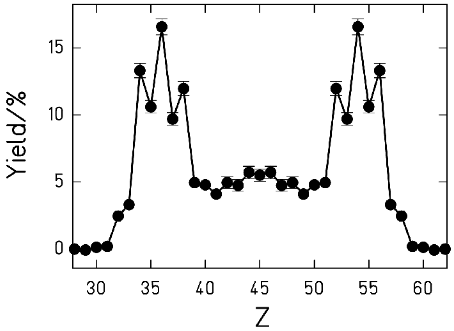

Fig. 1: Element distribution observed in the electromagnetic-induced fission of ^{229}Th (Schmidt et al., 2000).

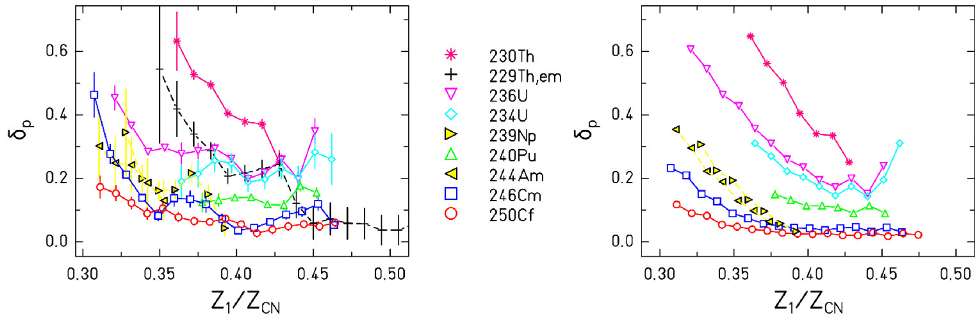

Fig. 2: Left part: Systematics of the measured local even-odd effect as a function of asymmetry, parameterised as the ratio of the Z of the light fragment and the Z of the fissioning nucleus, Z1/ZCN, for thermal-neutron-induced fission of heavy nuclei. The fissioning nucleus is indicated. The data have been taken from refs. (Rejmund et al., 2010; Caamano et al., 2011). The local even-odd effect of the electromagnetic-induced fission of ^{229}Th (see figure 1) is shown in addition. Right part: Result of a calculation with a schematic model (see text).

3. Previous explanations

The theoretical interpretation of the even-odd effect in fission-fragment yields was inspired for a long time by the observation that the magnitude of the effect is very sensitive to the initial excitation energy of the fissioning system and that no even-odd effect had been observed in odd-*Z* fissioning systems. Thus, the even-odd effect seemed to be a measure for the survival of a completely paired proton configuration at scission. Based on statistical concepts, several authors attempted to relate the magnitude of the even-odd structure in the *Z* yields with the intrinsic excitation energy available in the fissioning system in the vicinity of the scission point (Gönnenwein, 1991; Bocquet and Brissot, 1989). In this spirit, the lowering of the even-odd effect towards symmetry and the increase towards asymmetry was associated with "hot" symmetric fission and "cold" asymmetric fission (Nörenberg 1974). It seemed plausible that the amount of intrinsic excitation energy is reduced in very asymmetric fission due to the higher conditional fission barrier, since this interpretation is in line with the reduced yields. However, this explanation is not consistent with the assumption of "hot" symmetric fission, which is also characterized by low yields and a higher barrier in the heavier actinides (Vladuca et al., 2003).

Some attempts were made to theoretically study the dynamical process of pair breaking in the fission process (Bocquet and Brissot, 1989; Schütte, 1977; Bouzid et al, 1998; Krappe and Fadeev, 2001). But none of them can explain the drastic increase of the even-odd effect at large asymmetry. Qualitative arguments for this increase were given on the basis of the mass dependence of the pairing gap (Tsekhanovich et al, 1999) or the single-particle level density (Steinhäuser et al., 1998; Tsekhanovich, 2001), but they stayed on a purely statistical level, and the quantitative agreement with the data was not satisfactory.

4. Explanation by energy sorting

In the present work, we apply our recent considerations on the energy-sorting mechanism in fission dynamics under the strong influence of pairing interactions (Schmidt and Jurado, 2010) to propose a dynamical scenario for the asymmetry-associated even-odd effect in fission. It was shown that the constant-temperature behaviour of nuclei in the low-energy regime leads to an energy-sorting process if two nuclei are in thermal contact, as is the case in the fission process. Indeed, the level density ρ of a nucleus in the energy range of strong pairing interactions is rather well characterized by a constant-temperature behaviour (Schmidt and Jurado, 2012):

$$\rho \propto \exp\left(\frac{E^*}{T}\right) \tag{1}$$

with a temperature parameter T, which depends on mass number A and ground-state shell correction S, and, thus, is specific to the nucleus (von Egidy and Burescu, 2005):

$$T = A^{-\frac{2}{3}}\left(17.45 - 0.51\,S + 0.051\mathrm{S}^2\right) \tag{2}$$

Thus, in the absence of strong shell effects, the light fragment will have a higher temperature than the heavy one. Since the temperature of the fragments remains unchanged in spite of the variation of E^*, the light fragment will transfer essentially all its E^* to the heavy one (Schmidt and Jurado, 2011b). This process of energy sorting explains why an increase of the initial excitation energy leads to an increase of the number of emitted neutrons from the heavy fission fragment, only. This feature is observed in the excitation-energy dependence of the mass-dependent neutron yields. It seems natural and unavoidable that the complete energy sorting finally also favours the production of even-Z (and even-N) nuclides in the light fragment, because this leads to a considerable energy gain in the heavy fragment and, thus, to an increase in entropy of the system. The gain in E^* can be up to four times the pairing gap. Therefore, according to the energy-sorting mechanism, the hotter (normally the light) fragment tends to develop towards a fully paired configuration, i. e. with no quasi-particle excitations.

Let us now consider the dynamics of the energy-sorting process. The time t to form a fully paired light fragment is the sum of the time needed for the light fragment to transfer its E^* to the heavier one, and the time to exchange few nucleons through the neck. The latter time is rather short so that the time t is dominated by the time to transfer the E^* .

The energy transfer by unpaired nucleons has been considered, either the energy transfer between collective and intrinsic degrees of freedom (Randrup, 1979) or the transfer of entropy and intrinsic excitation energy between the fragments (Feldmeier, 1987; Schmidt and Jurado, 2011a). The contribution to energy transfer between the fragments due to the extension of the pairing correlations across the neck has been studied by solving the time-dependent pairing equations with constraint (Mirea, 2011). We limit ourself to a rough estimate of the exchange time of one unpaired nucleon. We assume a scenario with two fragments of A = 118 (radius 5.7 fm) connected by a neck with a typical radius of 3 fm. The nucleon moves with the Fermi velocity (83 fm / zs) inside one fragment, that means it hits the wall about 7.3 times per zs. The neck opens a window of 28 fm^2, which compares to the surface of 408 fm^2 of one fragment. Thus, the average time span between two transfers through the neck is about 2 zs. This estimate is also approximately valid for a typical mass-asymmetric fission. Inserting the typical energy transport of 0.5 MeV for one nucleon transfer (Schmidt and Jurado, 2011a), the energy flow by one nucleon amounts to 0.25 MeV / zs. Thus, it would require a typical time of 4 zs to transport an excitation energy of 1 MeV from the lighter to the heavier fragment. This may be compared to the typical dynamical saddle-to-scission time that amounts also to about 4 zs (Negele et al., 1978). Each quasi-particle excitation increases the number of unpaired nucleons by two, which tends to increase the excitation-energy flux through the neck accordingly. Thus, the time scale of the energy-sorting mechanism in low-energy fission, where excitation energies of a few MeV have to be transferred, is not very different from the dynamical saddle-to-scission time.

5. Schematical model

On the basis of this result, we develop a very simple schematic model of the even-odd effect in low-energy fission. We consider that the initial excitation energy $E^*_{0,light}$ is proportional to the available excitation energy at scission E^*_{sci} which is the sum of the excitation energy at saddle E^*_{CN} and the dissipated energy between saddle and scission $E^*_{sad-sci}$. E^*_{CN} increases with beam energy, and $E^*_{\mathrm{sad-sci}}$ increases with the Coulomb parameter $Z^2/A^{1/3}$ since the saddle to scission path becomes longer (Asghar and Hasse, 1984). On the other hand, the time t will decrease when the temperature difference $T_1 - T_2$ between the two fragments increases. A higher temperature gradient leads to faster flow of E^* between the two fragments. According to eq. (2). an increase in temperature difference corresponds to an increase in the asymmetry of the mass split. In a linear approximation, the time t follows the expression:

$$t \propto \frac{E^*_{sci}}{T_1 - T_2} \quad (3)$$

As a consequence, t will increase with the beam energy and the Coulomb parameter of the fissioning nucleus, and it will decrease with increasing asymmetry of the mass split. Eq. (3) is well reflected by the schematic drawing shown in Fig. 3, which illustrates the variation of the mean E^* in the light fragment as a function of time. Two fissioning nuclei and several mass splits, corresponding to equivalent mass asymmetries in both fissioning systems, are considered. One can see that the drop to $E^*=0$ (complete energy sorting) occurs faster for the more asymmetric splits. It also shows that the energy-sorting process takes longer for the heavier fissioning nucleus, because the E^* to be transferred is larger.

In the course of the motion of the fissioning system towards scission, there exists a time t_p , above which the exchange of protons through the neck is very much hindered due to the growing Coulomb barrier between the two fragments. If the time t required for complete energy sorting is larger than t_p , no net even-odd effect is induced because protons cannot be transferred through the neck. Thus, according to the energy-sorting process the even-odd effect as a function of asymmetry should have a threshold character. The threshold asymmetry where the even-odd effect created by the energy sorting sets in (corresponding to the asymmetry for which $t = t_p$) will increase with the Coulomb parameter of the fissioning nucleus. According to Fig. 3, in ^{236}U the energy sorting is accomplished within the time window t_p for the most asymmetric mass split (156/180) and, thus, the formation of an even-even light fragment is strongly enhanced. For ^{250}Cf, an even larger mass asymmetry than 165/85 is required. For a fixed even-Z fissioning nucleus, the general trend presented by the data in the left part of Fig. 2 is a small and rather constant even-odd effect close to symmetry and a strong increase as we move to more asymmetric fission. The latter feature occurs at an asymmetry value that increases with the mass of the fissioning nucleus, in agreement with what is expected from the energy-sorting process. For ^{230}Th, this change is not shown by the data. However, we presume that this is because the threshold asymmetry for this nucleus is close to symmetry where no data have been measured. The data of the electromagnetic-induced fission of ^{229}Th, which cover the whole mass range, support this assumption.

For several systems, the data point in Fig. 2 that is closest to symmetry is appreciably higher than expected from the global trend. This effect may be associated to the influence of the Z=50 shell in the complementary fragment, which is known to enhance the yield of tin isotopes and, thus, leads to a local increase of the magnitude of the even-odd effect deduced with the Tracy formula Tracy et al., 1972. In our schematic model, the dependence of the local even-odd effect with asymmetry is modelled in a phenomenological way with a step function, which is convoluted with a Gaussian function in order to account for some fluctuations in the dynamic evolution of the system. The threshold asymmetry value (the value at which the value of the smoothed step function is 1/2) is the one that fulfils the condition $C\,E^*_{sci}/|T_1-T_2| = t_p$, where C is a constant. t_p/C is adjusted to the data and has the same value for all nuclei. Since the energy sorting is not complete (Schmidt and Jurado, 2011b), the maximum possible even-odd effect is less than 100%. Best agreement with the data is obtained, if the width of the Gaussian function is set proportional to $|T_1 - T_2|$. The scaling factor for $|T_1 - T_2|$ is the same for all nuclei and fitted to the experimental data. In our model it is assumed that 30% of the energy release from saddle to scission (Asghar and Hasse, 1984) is dissipated into intrinsic excitations. The intrinsic excitation energy at scission determines also the magnitude of the odd-even effect at symmetry according to the model of ref. (Rejmund et al., 2000).

On the right part of Fig. 2, the results of the schematic model for the same fissioning systems are presented. The main tendencies of the experimental data are nicely reproduced by our description. The energy-sorting mechanism also predicts that, for a given fissioning nucleus, the threshold asymmetry should increase with increasing initial excitation energy of the compound nucleus. In addition, since the transfer of neutrons is possible until neck rupture, on expects smaller threshold asymmetries for the even-odd effect in the fission-fragment neutron yields. Unfortunately there are no data to verify these statements.

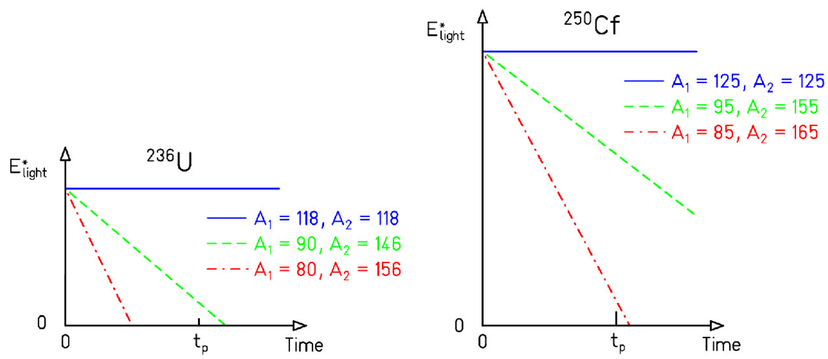

Fig. 3: Schematic drawing of the dynamical model based on the energy-sorting mechanism, which is proposed as the origin of the asymmetry-associated even-odd effect. The decrease of the excitation energy of the light fragment due to the energy-sorting mechanism is depicted for ^{236}U and ^{250}Cf and different mass splits. See text for details.

6. Conclusion

In conclusion, we state that the complex features of the even-odd effect in fission-fragment yields as a function of initial excitation energy and Coulomb parameter of the fissioning system as well as of the mass asymmetry of the fragments can easily be explained by the eventual transfer of the last unpaired proton from the light to the heavy fragment (or from the heavy to the light fragment) at the last step of the energy-sorting mechanism, which has recently been discovered. The fact that the even-odd effect is governed by the ratio of the total intrinsic excitation energy at scission and the temperature difference of the two nascent fragments fully agrees with the prediction of the model. Thus, the even-odd effect in fission-fragment yields gives a second strong evidence for the importance of the energy-sorting mechanism in low-energy fission which is fully consistent with the excitation-energy dependence of the prompt-fission neutron yields (Schmidt and Jurado, 2010).

This finding represents an essential progress in the understanding of fission dynamics: The threshold behaviour of the asymmetry-associated even-odd effect establishes a relation between the speed of the energy transfer in the energy-sorting mechanism and the dynamical time, starting at the moment when the two fragments develop their individual properties, e.g. their final temperatures, and the moment when the resistance against the transfer of protons across the neck becomes inhibitive. There exists some experimental knowledge on the saddle-to-scission time e.g. by the pre-scission neutron multiplicity at higher excitation energy (Hilscher and Rossner, 1992), but there is little knowledge on the time for intrinsic excitation-energy transfer between nuclei in thermal contact under the influence of pairing correlations. Detailed theoretical and experimental studies on pre-scission dynamics will allow extending the investigations on non-equilibrium processes between different super-fluid mesoscopic objects in analogy to the super-current (Peter et al., 2003) in particle transfer. In the present case, the driving force is the entropy, in contrast to transfer reactions, which are driven by different Fermi levels. Our finding provides an important constraint on the theoretical modelling of the last stage of fission under the influence of pairing correlations (Krappe and Fadeev, 2001), which represents still a considerable challenge. We expect that the progress in this field will finally provide valuable information for a better understanding of the microscopic nature of pairing correlations in nuclei.

The new insight into fission dynamics that has been made possible by the recent discovery of the energy-sorting mechanism reveals the wealth of information on the dynamical nuclear properties under the influence of pairing correlations, which is inherent in the complex features of even-odd structures in the fission observables. The theoretical tools for a quantitative interpretation on the microscopic level are still under development (Bulgac, 2010; Umar et al., 2010). Nevertheless, the schematic scenario presented in this letter gives convincing evidence that nature provides us with a very valuable rather direct information on several dynamical times. This finding will certainly stimulate the efforts for a better understanding of nuclear dynamics under the strong influence of pairing correlations.

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