GSI-Preprint-99-05
Januar 1999

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Pair Breaking and Even-Odd Structure in Fission-Fragment Yields

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Abstract The probabilities of single-particle excitations which preserve completely paired configurations of the nuclear proton and neutron subsystems are formulated as a function of excitation energy in the framework of the superfluid nuclear model. These calculations are used to analyse measured data on even-odd structure in fission-fragment yields. Excitation energies acquired at scission are deduced. The strongly differing even-odd structures in proton and neutron numbers are explained within the assumption of thermal equilibrium of intrinsic excitations at scission.

PACS: 25.85.-w; 24.10.Pa; 21.10.Ma

Keywords: Nuclear fission; Pair breaking; Even-odd structure in fission-fragment yields; Statistical model; Superfluid nuclear model

Introduction

Viscosity, that means the coupling between collective motion and the intrinsic single-particle degrees of freedom, is one of the basic nuclear properties that is presently under intensive investigation. For these studies, the fission process is one of the most important sources of information. While the viscosity of hot nuclear matter is determined from the time scale of high-energy fission [1], the viscosity of cold nuclear matter is deduced from the even-odd structure observed in fission-fragment yields after low-energy fission [2]. Motivated by the recent experimental progress brought about by the use of secondary beams, we will reconsider the theoretical understanding of the onset of dissipation in a completely condensed microscopic system due to a large-scale collective motion. The present paper revisits the relation between the even-odd structure and pair breaking in fission on the basis of the statistical model.

The enhanced production of nuclei with even numbers of protons and neutrons is one of the prominent structural effects in nuclear fission from low excitation energies [3]. However, the experimental access to the exact number of nucleons of the fission fragments directly after scission is difficult to obtain. Since the number of neutrons is modified by evaporation processes, only in cold fission, i. e. fission with kinetic energies close to the Q value (the total energy released), the number of neutrons observed represents the situation at scission. On the other hand, evaporation of protons from the neutron-rich fission fragments is very much suppressed. Therefore, the element yields produced at scission may be obtained, if the detection method is faster than beta decay. This criterion is met e. g. by experiments at Lohengrin [4,5,6,7,8,9,10] and Cobi Fan Tutte [11,12,13,14] at the ILL neutron high-flux

reactor in Grenoble. In all these experiments, fission was induced by the capture of thermal neutrons. Therefore, the excitation energy of the fissioning nuclei could not be varied. Results on the excitation-energy dependence of proton even-odd effects in photo-induced fission have recently been deduced [15,16] from gamma-spectroscopic studies.

The total even-odd effect in element yields, defined as the difference of even-Z and odd-Z yields divided by the total yield, has been found to vary systematically as a function of fissility of the fissioning nucleus. Values ranging from 41% for ^{230}Th to 4.6% for ^{250}Cf have been observed in thermal-neutron-induced fission [2]. Although the number of neutrons of the fission fragments produced at scission is difficult to measure, data from high kinetic energies where neutron evaporation is suppressed show that the even-odd effect in neutron number is considerably reduced as compared to the even-odd effect in proton number [2].

In an experiment recently performed at GSI Darmstadt with relativistic secondary heavy-ion beams, element yields have been determined for long isotopic series of neutron-deficient actinides and pre-actinides [17,18]. 70 different isotopes were produced as secondary projectiles and excited by electromagnetic interactions in a lead target, leading to fission from excitation energies around 11 MeV. For the first time, the systematic variation of the proton even-odd effect as a function of neutron and proton number of the fissioning system was measured over a large region of the chart of the nuclides. Due to the considerably increased data basis on even-odd effects in fission, the systematic appearance of an even-odd structure also in odd-Z fissioning nuclei has been revealed. In addition, the data show that an increased even-odd structure in extremely asymmetric charge splits is a general effect for even-Z fissioning nuclei. These new findings illustrate the perspectives for a new insight into the mechanism of pair breaking in fission brought about by the progress in experimental technique.

Several models have been proposed to quantitatively relate the observed enhancement of even elements in fission-fragment Z distributions to the intrinsic excitation energy at scission either on a combinatorial basis [19] or with a thermodynamical approach [20]. However, none of them is able to reproduce the systematic appearance of even-odd structures in odd-Z fissioning nuclei [18] or the observed differences of the even-odd effects in proton and neutron number [2] within the assumption of a thermally equilibrated scission-point configuration.

In the present paper, we deduce the survival probability of the completely paired proton or neutron subsystems from the statistical weight of the corresponding excited nuclear states in the frame of the superfluid nuclear model. Such an approach differs conceptionally from the other previously proposed models. We think that the treatment presented here corresponds in the most direct way to the fundamental concepts of the statistical model.

Formulation of the Model

One important step on the way to a quantitative formulation of the problem is the description of quasi-particle excitations in the transitional saddle configuration of the fissioning nucleus. On the way from the ground-state deformation to the fission barrier, the nucleus has to transform the necessary amount of energy from intrinsic excitations into the deformation degree of freedom in order to overcome the barrier. Therefore, the intrinsic excitation energy decreases on the way to the barrier. In many cases, e.g. in thermal-neutron-induced fission, only completely paired transition states are available. The single-particle excitations in the transition states at the fission barrier represent the starting conditions for the dynamical evolution towards scission. From saddle to scission, the coupling between

collective and single-particle degrees of freedom introduces an additional intrinsic excitation energy to the system which leads to the breaking of pairs.

Experimental data on the even-odd effects in the fission-fragment yields [5-10,12-14] and most of all the stabilisation of such effects for excitation energies lower than $2\Delta_0$ above the fission barriers [15,16] lead us to suppose that the observed proton even-odd effects are strongly connected with the maximum paired proton configurations. While it is sufficient to introduce an excitation energy which exceeds the pairing gap $2\Delta_0$ in order to break at least one pair and thus to destroy the completely paired configuration, this is not true if only the proton or neutron subsystem is considered. Up to rather high excitation energies, there is a considerable probability that the intrinsic excitation energy is restricted to protons or neutrons, respectively, and that a completely paired configuration of the neutron or proton subsystem, respectively, is preserved. This gives rise to a proton even-odd effect, even though the excitation energy is sufficient to break one or several pairs. The same is true for the neutron even-odd effect.

The survival probability of the completely paired proton configuration ($n_Z=0$) in an excited nucleus can be defined on the basis of the statistical consideration as the ratio of the corresponding level densities:

$$P_0^Z(U) = \sum_{n_N} \rho_{n_Z=0, n_N}(U) / \sum_{n_Z, n_N} \rho_{n_Z, n_N}(U) , \quad (1)$$

where $\rho_{n_Z, n_N}(U)$ is the level density of n_Z -proton and n_N -neutron quasiparticle excitations at the given energy U . The sum in the numerator is taken over all possible neutron excitations, and the sum in the denominator includes also all possible proton excitations. It is obvious that the last sum coincides with the total level density of the considered nucleus.

The main features of the present model, primarily intended to describe the pairing correlations at the fission barrier, may also be applied to the scission configuration. If we assume that a full dynamical treatment is only necessary to deduce the amount of dissipated energy on the way to scission, the application of the statistical model is justified to formulate the probabilities of competing single-particle configurations at scission, too.

Pairing effects in the nuclear level density

The main properties of the level density at the saddle point and in all following stages between saddle and scission differ only quantitatively from the regularities of the level-density behaviour in the ground-state configurations of nuclei. Therefore, we will first discuss the general features of the pairing effects in the nuclear level density.

The consideration of the statistical properties of nuclear excitations with a fixed number n of excited quasiparticles can be performed on the basis of a rather simple formula for the density of such excitations firstly proposed by Strutinsky [21]:

$$\rho_n(U) = \frac{g^n (U - n\Delta)^{n-1}}{[(n/2)!]^2 (n-1)!} , \quad (2)$$

where U is the excitation energy, g is the density of single-particle states near the Fermi energy, and Δ is the pairing-gap parameter.

Ignatyuk et al. [22,23] proposed a more consistent description of the statistics of nuclear excitations based on the superfluid nuclear model. In this approach, the correlation function Δ_n depends on both the number of excited quasiparticles and excitation energy, and the level density may be written in the form of

$$\rho_n(U) = \frac{g^n}{[(n/2)!]^2 (n-1)!} \left[U - \frac{1}{4} g (\Delta_0^2 - \Delta_n^2) - \Gamma_n \right]^{n-1}, \quad (3)$$

where Δ_0 is the correlation function or pairing-gap parameter for the ground state of the nucleus and Γ_n is the correction that takes into account the influence of the Pauli exclusion principle on nuclear excitations [24]. The dependence of Δ_n upon the excitation energy should be obtained from the corresponding state equations of an excited nucleus, the general solutions of which were considered in Ref. [22].

A rather simple analytical parameterisation of these solutions was proposed by Fu [25], who also discussed an application of the model for a two-component system [26]. Comparing the parameterisation [25] with the exact solutions of the superfluid-model equations, we have found that the accuracy of the parameterised solutions can be increased if we define some of the numerical coefficients in them on the basis of the analytical expressions obtained in [22]. Taking into account these modifications, the threshold energy U_{th} , above which n -quasiparticle excitations can exist, may be defined by the following expressions:

$$\frac{U_{th}}{C} = 3.144(n/n_c) - 1.234(n/n_c)^2 \quad \text{for } n/n_c \leq 0.424 \quad (4)$$

$$\frac{U_{th}}{C} = 1 + 0.617(n/n_c)^2 \quad \text{for } n/n_c > 0.424$$

Here $C = g\Delta_0^2/4$ is the condensation energy and $n_c = 0.791g\Delta_0$ is the number of excited quasiparticles at the phase-transition point from the superfluid state to the normal one (a full discussion of the physical meaning of these nuclear characteristics may be found in Ref. [22]). In the superfluid model, the threshold for the excitation of two quasiparticles does not coincide exactly with $2\Delta_0$.

The energy dependence of the pairing-gap parameter, that is required for calculations of the level density (3), can be parameterised in the form:

$$\frac{\Delta_n}{\Delta_0} = 0.996 - 2.36(n/n_c)^{1.57} / (U/C)^{0.76} \quad \text{for } \frac{U}{C} \geq 1.03 + 2.07(n/n_c)^{2.91}, \quad (5)$$

$$\frac{\Delta_n}{\Delta_0} = 0 \quad \text{otherwise.}$$

Eqs. (4) and (5) differ only by the coefficients from the similar expressions of Fu [25]. The difference between our parameterisation and that of Fu is rather small and not essential for the following consideration.

Main parameters

The energy dependencies of the correlation functions and the level densities calculated on the basis of Eqs. (3-5) for different numbers of excited proton or neutron quasiparticles in ^{232}Th are shown in Fig. 1. For these calculations we use the correlation functions applied in the recent mass formula [27]

$$\Delta_{0\tau} = 3.2 / N_r^{1/3} \text{ MeV}, \quad (6)$$

Fig. 1. Dependence of the level densities (upper part) and correlation functions (lower part) of proton or neutron excitations in ^{232}Th with a given number of excited quasiparticles on the excitation energy.

and the single-particle level densities

$$g_{\tau} = N_{\tau} / 15 \text{ MeV}^{-1}. \quad (7)$$

Here N_{τ} is the number of protons ($\tau = Z$) or neutrons ($\tau = N$), respectively. The decrease of the correlation functions with increasing number of excited quasiparticles is a very important effect that leads to essential differences of the nuclear level densities, Eq. (3), from the Boltzmann-gas description with an unchanged pairing-gap parameter, Eq. (2). Excitations with the number of quasiparticles $n > n_c$ have correlation functions equal to zero at the near-threshold region, and the pairing correlations arise at higher excitations only when excited particles are distributed far away from the Fermi level.

The expansion of the considered equations to a two-component system of protons and neutrons is rather simple. The ground-state correlation functions $\Delta_{0\tau}$ and the single-particle level densities g_{τ} should be specified for protons and neutrons, then $n_{c\tau}$ and C_{τ} should be calculated, and U_{th} must be estimated from Eq. (4) for each component. Finally, the level density of excitations with a definite number of excited proton and neutron quasiparticles may be calculated from the equations similar to Eq. (3). The general forms of the two-component expressions for the level densities and the Pauli corrections are well known [28], and the main details of their application to our task are presented in Appendix 1.

To understand the influence of single-particle level densities on the even-odd effects analysed, we also used in the following calculations the single-particle densities defined as

$$g_{\tau} = \frac{N_{\tau}^{1/3} A^{2/3}}{2^{2/3} 15} \text{ MeV}^{-1}. \quad (8)$$

This formula separately takes into account the influence of the number of particles on the nuclear volume and on the chemical potential. It corresponds to the semi-classical estimation of the single-particle densities of the two-component Fermi gas, which are considered in Appendix 2. Eq. (8) gives almost the same total level density $g = g_Z + g_N$ as the total level density determined by Eq. (7), but a different contribution of each component.

Situation at the fission barrier

In order to represent the situation at the fission barrier in a realistic way, the deformation dependence of the pairing gap should be taken into account. From the analysis of available experimental data on the fission cross sections at near-threshold energies [29] it follows that the pairing-gap parameters at the saddle have to be taken 15% higher than for ground-state deformations. In accordance with these results, we increased the numerical coefficients of Eq. (6) by 15% in calculations of the level densities in the saddle configuration. For the single-particle level densities defined by Eq.(7), the resulting calculations of the survival probability of a completely paired proton configuration ($n_Z=0$) are shown in Fig. 2a. The narrow slit of $P_0^Z(U)$ near the threshold of the two-quasiparticle excitation is a direct consequence of the lower threshold of proton excitations relative to the neutron-excitation threshold.

Calculations of the survival probabilities for the single-particle level densities defined by Eq. (8) are shown in Fig. 2a, too. Due to the modified threshold energies according to Eq. (4),

the slit of $P_0^Z(U)$ at the two-quasiparticle threshold disappeared, but other regularities of the energy behaviour of $P_0^Z(U)$ remained rather similar.

In the same way one can define the survival probability $P_0^N(U)$ of the completely paired neutron configuration ($n_N = 0$). In this case, the sum in the numerator of Eq. (1) must be taken over all possible proton excitations. The results of these calculations for the two considered sets of the single-particle level-density parameters are shown in Fig. 2b. The survival probability of paired neutron configurations decreases much faster than the probability for paired protons. This property is a direct consequence of the higher contribution of neutron excitations to the total level density.

Fig. 2. Dependence of the survival probabilities of completely paired proton (a) and neutron (b) configurations in ^{232}Th on the excitation energy above the saddle. The solid and dashed curves correspond to Eqs. (7) and (8), respectively, for the single-particle level densities. For excitation energies below the lowest threshold for quasiparticle excitations at the saddle, fission proceeds by the completely paired single-particle configuration.

Discussion of the results

Energy dependence of the survival probability at the saddle point

The quantitative predictions of our model on excitation-energy dependent even-odd structure in the fission-fragment charge yields may be compared with the data recently

reported for two fissioning systems: ^{238}U [15] and ^{232}Th [16]. In order to allow for this comparison, an assumption has to be made on the variation of the dissipation from saddle to scission as a function of intrinsic excitation energy at scission. As a first approximation we might assume that the influence of the whole path from saddle to scission corresponds to some additional dissipation that is independent from the initial excitation conditions at the saddle point. This assumption is used now practically in all analyses of observed even-odd effects [2]. Of course, we are aware that this could be an oversimplification since it is to be expected that a completely paired system which can only be excited to energies exceeding the pairing gap has a lower viscosity than an excited system which can absorb much lower amounts of energy [30].

Fig. 3. Comparison of the calculated survival probabilities P_0^Z at scission of the completely paired protons (full lines) with experimental data on the even-odd differences δ_Z of fission-fragment element yields for the fissioning nuclei ^{238}U (triangles), and ^{236}U (squares), and ^{232}Th (crosses). The data are taken from refs. [15,16]. In addition, the dotted curves represent the empirical parameterisations proposed in refs. [15,16].

In accordance with this approximation we multiplied the proton survival probabilities with a coefficient of 0.3 determined from the observed even-odd effects for the excitation energy at saddle equal to zero [15,16]. Results of such calculations are shown in Fig. 3 together with available experimental data and the empirical curve defined in Ref. [15]. Here and in the following, the single-particle level densities according to Eq. (8) are used. The main feature of

the analytical approximation proposed in Refs. [15,16] that the even-odd effect persists up to an energy of about $2\Delta_0$ is in accordance with our model. Our predictions for the energy dependence of the proton even-odd effect are almost identical for the two nuclei. According to our model, the even-odd effect decreases more gradually than the experimental data. The steeper descent of the experimental even-odd effect might comply with the expectation [30] that nuclear viscosity increases with temperature. In view of the experimental uncertainties, however, any final conclusion cannot yet be drawn.

Dissipation from saddle to scission

In accordance with experimental data available, the even-odd structure in fission-fragment yields depends on both the excitation energy at saddle and the dissipated energy on the path from saddle to scission. A possible influence of saddle configurations was considered above. It is natural to assume that the survival probability can be used to describe the even-odd effects on all post-saddle stages, too. To get the total effect, it is necessary to write and solve the corresponding dynamical equations for the survival probability changes. Such a task is rather complex.

However, we can assume that a contribution of all post-saddle stages may be approximated by the survival-probability description at some effective scission point. Of course, such a point differs from the geometrical scission configuration and relates to an earlier stage of fission. In accordance with this assumption the total survival probability of paired proton configurations should be defined as the product of the corresponding probabilities at saddle and scission.

The differences between the survival probabilities at saddle and scission are determined by possible changes of the correlation functions Δ_r . These functions have larger values in the saddle configuration than in the ground state, and we should await some additional increase of them for the more deformed scission configuration. It seems natural to assume that the correlation functions at scission are approximately equal to their values in the separated fragments. However, if one tries to calculate them with Eq. (6) the values obtained are appreciably lower than the experimental pairing corrections for the mass region of $A \approx 115$ [31, 32]. It seems that Eq. (6) correctly reproduces the difference between the proton and neutron correlation functions of heavy nuclei, but underestimates their absolute values for medium and light nuclei. To correct this shortcoming, the mass dependence of pairing corrections with $1/\sqrt{A}$ recommended in Ref. [31] was applied, and in accordance with it the ratio $\Delta_{scis}/\Delta_0 = \sqrt{2}$ was used to estimate the increase of the correlation functions at scission. The survival probabilities of the completely paired proton and neutron configurations calculated for the corresponding values of Δ_Z and Δ_N are shown in Fig. 4. Single-particle level densities according to Eq. (8) were used. The increase of the correlation functions reduces the damping of the survival probability at scission relative to the damping of the survival probability at saddle (compare with Fig. 2a). The experimental data presented in Fig.4 will be discussed later.

The potential-energy gain between saddle and scission increases with increasing Coulomb parameter $Z^2/A^{1/3}$ of the fissioning nucleus [33]. It has always been a challenge to estimate the fraction of this energy gain which is transformed into the intrinsic excitation energy in order to obtain information on the viscosity of nuclear matter. Such an analysis has been performed by Gönnerwein [2] on the basis of the combinatorial model [19]. A simple formula connecting the even-odd differences of fission-fragment element yields δ_Z with the dissipation or intrinsic excitation energy was obtained:

$$E_{dis} = -4 \ln \delta_Z \quad (9)$$

Available experimental data from thermal-neutron induced fission on the total even-odd effects are given in Fig. 5, upper part, as a function of the Coulomb parameter $Z^2/A^{1/3}$. With the exception of ^{250}Cf , these nuclei had to pass through completely paired transition-state configurations. The intrinsic excitation energies acquired at scission as deduced on the basis of Eq. (9) are shown in Fig. 5, central part, in comparison with the calculated energy release from saddle to scission [34]. It was concluded from these results that an almost constant fraction of about 30% of the energy release is absorbed into intrinsic excitations (see Fig. 5, lower part).

Fig 4. Dependence of survival probabilities P_0^Z (full line) and P_0^N (dashed line) of the completely paired proton and neutron configurations on the excitation energy at the effective scission point. The experimental data on the proton and neutron even-odd effects δ_Z and δ_N at fixed kinetic energies of the light fission fragments are shown for the fissioning nuclei ^{234}U ($E_{\text{kin}} = 111$ MeV), ^{236}U ($E_{\text{kin}} = 108$ MeV), and ^{240}Pu ($E_{\text{kin}} = 111$ MeV) by closed and open symbols, respectively.

It seems to be adequate to repeat this analysis on the basis of the statistical considerations of the present work. However, at first we should take into account the asymmetry contribution to the even-odd effect.

It was demonstrated in the GSI experiments [18], that the local even-odd effect $\delta_Z(Z)$ [35] which was observed for the whole range of the charge-yield distributions, increases for asymmetric charge splits with respect to the even-odd effect in symmetric fission. This effect was explained on the basis of the phase space available in the two nascent fragments for unpaired protons [18]. The relative statistical weight at scission of configurations with one unpaired proton in one fragment is determined by the probability

$$p(Z) = \frac{g(Z)}{g(Z) + g(Z_f - Z)} = \frac{Z}{Z_f} \quad (10)$$

Fig. 5. Analysis of even-odd effects in the nuclear charge yields observed in thermal-neutron-induced fission of different nuclei (open and full symbols) [5-14] and in electromagnetic-induced fission of $^{220-228}\text{Th}$ (crosses) [18].

Upper part: Even-odd effects in nuclear charge yields. *Open symbols:* Measured total even-odd effect, *Full symbols:* Same data, corrected for the asymmetry effect (see text). *Crosses:* Measured local even-odd effects in near-symmetric charge splits for thorium isotopes after electromagnetic-induced fission.

Central part: Excitation energies at scission, deduced from the corresponding data points in the upper part of this figure. *Open symbols:* Excitation energies at scission, deduced from the total even-odd effects with Eq. (9). *Full symbols and crosses:* Excitation energies at scission, deduced from the corrected even-odd effects with the full curve of Fig. 4. *Asterisks:* Energy release from saddle to scission [34].

Lower part: Fraction of the energy release, which is dissipated on the way from saddle to scission. Data symbols correspond to those used in the central part of the figure.

The dashed lines are drawn to show the trends of our analysis.

where Z_f is the nuclear charge of the fissioning nucleus. In accordance with the combinatorial consideration of the phase space available, the resulting local proton even-odd effect in the vicinity of the fission-fragment charge Z for an even- Z fissioning nucleus is described by the relation

$$\delta_z(Z) = \sum_{n=0,2} P_n^Z \cdot [1 - 2p(Z)]^n \quad . \quad (11)$$

We remind that P_n^Z denotes the probability for n -proton quasiparticle excitations. In Eq. (11) we consider only the completely paired and the two-quasiparticle states. In this consideration, contributions to the local even-odd effect with $n > 0$ vanish in symmetric fission, and the observed odd- Z fragment yields at symmetry are a direct measure of the breaking of nucleon pairs due to the dissipation from saddle to scission. A detailed discussion of the relation between the local and the total even-odd effects and of the influence of phase space on the even-odd structure is given in ref. [18].

To get a realistic estimation of the dissipated energy, it is necessary to determine the survival probability P_0^Z of the completely paired configuration. We considered the thermal-neutron-induced fission of ^{233}U [9], ^{235}U [7], ^{239}Pu [8], ^{241}Pu [14], and ^{249}Cf [10]. For each fissioning system, P_0^Z and $P_2^Z = 1 - P_0^Z$ were adjusted to reproduce the measured total even-odd effect δ_z by the local even-odd effect $\delta_z(Z)$ with Eq. (12) for the average charge in the asymmetric part of the Z distribution. The correction might be slightly overestimated, since contributions with $n > 2$ were neglected.

The even-odd effect at symmetry obtained after taking into account the corrections considered is shown in the upper part of Fig. 5 together with the original experimental data. In the central part of Fig. 5, the intrinsic excitation energies obtained for these values of δ_z from the solid curve of Fig. 4 are presented. The decrease of the correlation functions Δ_r from ^{232}Th to ^{250}Cf is rather small, and the application of the same curve for all considered nuclei is a good approximation. It can be seen that after correction of the asymmetry effect and the application of the new model the absolute values are rather close to those obtained by Gönnerwein [2]. However, the increase as a function of the Coulomb parameter $Z^2/A^{1/3}$ is found to be smaller. As a result, the dissipated fraction of the energy release between saddle and scission is decreasing with increasing Coulomb parameter (see lower part of Fig. 5).

Different even-odd effects for protons and neutrons

Above, the quantitative expressions for two groups of single-particle configurations were considered, first those excitations which preserve the completely paired proton configuration and which allow only for quasiparticle excitations in the neutron subsystem, secondly those which preserve the completely paired neutron configuration and which allow only for quasiparticle excitations in the proton subsystem. In our model, just these configurations are responsible for the even-odd difference of observed fission-fragment yields. With increasing excitation energy, both paired configurations represent a decreasing fraction of all possible nuclear excitations, and thus both the neutron and the proton even-odd effects decrease with increasing energy. The two expressions look identical but the numerical results differ considerably, because in heavy nuclei the number of neutrons exceeds that of the protons by about a factor of 1.5. Since the single-particle level density of the neutrons exceeds that of the protons, the nuclear level densities of the neutron subsystem grows more strongly with increasing number of quasiparticle excitations than the nuclear level density of the proton subsystem according to Eq. (3). This is why the statistical weight of pure neutron excitations

($n_Z=0$) is appreciably larger than that of pure proton excitations ($n_N=0$). As a consequence, the proton even-odd effect should decrease more gradually with increasing energy than the neutron even-odd effect does.

Fission at moderate excitation energies above the pairing gap but with pairing correlations still preserved is thus expected to show appreciably stronger even-odd effects in proton number than in neutron number. Such a difference in the experimental data, even at high kinetic energies where neutron evaporation cannot occur, has always been a puzzle [2]. Since previous statistical considerations did not give a quantitative explanation, one was obliged to attribute this effect to the fission dynamics. As a possible explanation, one might assume that the protons are separated in an earlier stage than the neutrons due to Coulomb repulsion. Thus, for the neutron pairs an additional probability to be separated in the most violent dynamics of the neck rupture was expected. The statistical description proposed in the present work, however, explains this difference without considering any difference in the dynamics of protons and neutrons at scission.

The isotopic and isotonic yields of fission fragments from the thermal-neutron-induced fission of ^{233}U , ^{235}U , and ^{239}Pu were experimentally investigated [7-9]. Since the even-odd effect for isotonic yields is strongly influenced by neutron evaporation, valuable information on this quantity is obtained only for the highest kinetic energies of the fragments, which corresponds to the coldest fission processes where no neutron evaporation is possible. According to the present model, each measured even-odd effect in element yields may be associated with one specific intrinsic excitation energy. Considering the results based on the single-particle level density described by Eq. (8), the excitation energies corresponding to the observed even-odd proton effects were estimated. These data are shown in Fig. 4. The calculated even-odd effects in isotonic yields for these energies are in remarkable agreement with the experimentally observed values for ^{233}U and ^{235}U .

The data for ^{239}Pu are not so well described by our model. For this nucleus, the even-odd effect in neutron yields is half as large as that in element yields. Unfortunately, we could not extend this analysis to other systems because to our knowledge there exist no other data on neutron even-odd structure at high kinetic energies.

The data presented on Fig. 4 were not corrected for the asymmetry contribution to the even-odd effect. However, as it can be seen from Fig. 5, the corrections are not very important and would not lead to different conclusions.

GSI data

The experiments performed at GSI with secondary beams of relativistic heavy ions have provided data on the local proton even-odd effect for long isotopic chains [18]. For the first time, the even-odd effect has systematically been studied not only for asymmetric but also for symmetric charge splits. The data at symmetry are particularly interesting since here the even-odd structure in element yields represents the probability for proton quasiparticle excitations without any correction needed and the data can directly be interpreted with our model. In the present paper we restrict our analysis to electromagnetic-induced fission of the thorium isotopes ^{220}Th to ^{228}Th . For all these isotopes, a value of δ_Z around 0.09 has been obtained for symmetric charge splits [18].

These data correspond to excitation energies of the compound nuclei around 11 MeV [36] and to an energy excess above the fission barrier of about 4.5 MeV. From a comparison with the theoretical curves in Fig. 4 we conclude that the measured value of $\delta_Z = 0.09$ corresponds to excitation energies at scission between 8 and 9 MeV. Thus, the gain of excitation energy due to dissipation from saddle to scission amounts to 3.5 - 4.5 MeV. These values are close to

the dissipated energies obtained from available data on thermal-neutron induced fission of thorium isotopes (see Fig. 5, central part). This result gives no indication for an increased dissipation when the system starts from higher excitation energies.

These data allow us to comment on the appropriate ordering parameter for the dissipated energy on the way from saddle to scission. Previous data are described equally well as a function of the fissility parameter Z^2/A [2] and as a function of the Coulomb parameter $Z^2/A^{1/3}$ [37]. The fact that no significant variation of the dissipated energy is observed over 9 thorium isotopes proves that the influence of mass number is relatively small. Therefore, the Coulomb parameter seems to be best adapted for the parameterisation of dissipation effects in fission. This is why $Z^2/A^{1/3}$ was chosen as the ordering parameter in Fig. 5.

It should be noted that the simple estimations considered above are made for the average excitation energy of the electromagnetic-induced fission. However, the distribution of compound-nucleus energies after electromagnetic excitation is rather broad, and the high-energy tail of the distribution should be taken into account for a more consistent comparison of theory and experiment. Such an analysis requires a special consideration that lies out of the scope of the present paper.

Comparison with previously proposed models

Finally, we would like to outline the fundamental differences of our statistical considerations to the expectations of previously proposed models. Let us remind that the present model takes the number of nuclear excitations available at a given excitation energy as the decisive quantity for the statistical considerations, see Eq. (1). The probability for completely paired configurations of the proton nuclear subsystem is expressed as the relative statistical weight of those excitations which are restricted to neutrons only and hence do not involve any protons. If such a configuration is realised at scission, only fission fragments with even proton numbers can be formed.

First, the combinatorial model of Nifenecker et al. [19] is characterised by a fundamental difference to our approach: It considers the number of nucleon pairs as a statistical unit for combinatorial considerations.

Secondly, the thermodynamical model of Mantzouranis and Nix [20] considers the number of quasiparticles above the Fermi level as the key quantity for the even-odd effect. The proton even-odd effect persists even when proton pairs are broken. It disappears at the critical temperature, only. In contrast to our model, the even-odd effects in proton and neutron number are identical at a given temperature when the pairing gaps are equal, independent of the N/Z ratio of the fissioning nucleus.

In view of these fundamental differences it seems fortuitous that the quantitative predictions of all three models on the excitation-energy dependence of the even-odd effect are not very different quantitatively. However, there is a decisive discrepancy in the comparison of the even-odd effects in neutron and proton number. Only our approach predicts an appreciable difference, which is in quantitative agreement with the characteristics of experimental data.

Conclusion

A model has been formulated for the interpretation of the even-odd structure in fission fragment yields. The model is based on the number of available single-particle excitations of the nuclear system, calculated in the framework of the superfluid nuclear model. Assuming thermal equilibrium of the single-particle excitations, the probability of pair breaking is given

by the relative statistical weight of excitations which destroy the completely paired configuration of the proton, respectively neutron subsystem at the different stages of the fission process. The concept of the new model differs appreciably from that of previously proposed ones. Our approach seems to be more consistent with the basic principle of the statistical theory of nuclear reactions.

From the even-odd structure of measured fission-fragment element yields, the excitation energy acquired at scission was deduced. Contributions to the even-odd structure from the different phase spaces in the nascent fission fragments were considered. Indications have been found that the energy dissipation in fission scales with the Coulomb parameter $Z^2/A^{1/3}$ rather than with the fissility of the fissioning system. The deduced excitation energies at scission were found to increase less steeply with the Coulomb parameter of the fissioning nucleus than deduced previously. From the available data on even-odd effects in fission from higher excitation energies, no clear indication for a temperature dependence of the nuclear viscosity could be deduced.

The model explains the experimental observation that neutron even-odd effects, even in cold fission, are considerably smaller than proton even-odd effects without invoking different influences of nuclear dynamics on the proton and neutron subsystems. The strongly different even-odd structures in proton and neutron number well comply with the assumption of thermal equilibrium in the scission configuration.

Acknowledgement

This work has been supported by the Human Capital and Mobility Program of the European Union and by the WTZ Program of the Bundesministerium für Bildung, Wissenschaft, Forschung und Technologie. The responsibility rests with the authors.

Appendix 1

The formulae for the level density of quasiparticle excitations in a two-component system of protons and neutrons differ from the one-component relation, Eq. (2) or (3), only by the factorials in the denominators and a more complex form of the Pauli corrections [28]. They may be written for an even number of protons in the form of

$$\rho_{n_Z, n_N}(U) = \frac{(g_Z^{n_Z} + g_N^{n_N}) \left[U - \sum_{\tau} (P_{n_{\tau}}^{pair} + \Pi_{n_{\tau}}) \right]^{n_Z + n_N - 1}}{(n_Z + n_N - 1) [(n_Z / 2)!]^2} \times \begin{cases} [(n_N / 2)!]^2 \\ [((n_N + 1) / 2)!]^{-1} [((n_N - 1) / 2)!]^{-1} \end{cases} \quad (A1.1)$$

The upper option corresponds to nuclei with an even and the lower to those with an odd number of neutrons. $P_{n_{\tau}}^{pair} = g_{\tau} [\Delta_{0\tau}^2 - \Delta_{n_{\tau}}^2(U_{\tau})] / 4$ is the energy-dependent pairing correction and $\Pi_{n_{\tau}}$ is the correction taking into account the Pauli exclusion principle. These corrections are independent for protons and neutrons. The changes of the factorials to be made for odd- Z nuclei are obvious from the view of the last multipliers of Eq. (A1.1).

The existence region and the value of $\Delta_{n_{\tau}}(U_{\tau})$ are determined by Eqs. (4) and (5) for the given excitation energy U_{τ} of each component. The excitation energy is proportional to the average number of excited quasiparticles and this permits to use a rather simple split of the full excitation energy on the proton and neutron components:

$$U_{\tau} = \frac{n_{\tau}}{n_Z + n_N} U \quad . \quad (A1.2)$$

A more rigorous analysis of the energy split on the basis of the exact state equations of the superfluid model [22] confirms a good accuracy of the approximation (A1.2). It is necessary to note that for odd N_{τ} the ground-state value of $\Delta_{0\tau}$ should be calculated as the lowest one-quasiparticle excitation.

The Pauli corrections are energy independent and defined as

$$\Pi_{\tau} = \begin{cases} n_{\tau}(n_{\tau} - 2) / 8g_{\tau} & \text{for even } N_{\tau}, \\ [(n_{\tau} - 1)^2 + 4] / 8g_{\tau} & \text{for odd } N_{\tau}. \end{cases} \quad (A1.3)$$

Appendix 2

A simple estimation of the single-particle level density may be obtained on the basis of the semiclassical approximation [23]. The Bohr-Sommerfeld conditions determining the energies ϵ of single-particle levels can be written in this approach for a spherical potential square well as

$$\pi(n+1/2) = \int_{r_{\min}}^{R_0} \left[\frac{2\mu\epsilon}{\hbar^2} - \frac{(l+1/2)^2}{r^2} \right]^{1/2} dr, \quad (\text{A2.1})$$

where μ is the nucleon mass, l is the orbital angular momentum and $R_0 = r_0 A^{1/3}$ is the nuclear radius. The density of single-particle states with a given value of the angular momentum is defined for the proton or neutron subsystem ($\tau = Z$ or N) by the relation

$$g_\tau(\epsilon, l) = 2(2l+1)dn_\tau / d\epsilon. \quad (\text{A2.1})$$

One can obtain the total single-particle state density by calculating the corresponding integrals

$$g_\tau(\epsilon) = \int_0^{l_{\max}} g_\tau(\epsilon, l) dl = \left(\frac{2\mu R_0^2}{\hbar^2} \right)^{3/2} \frac{2\epsilon^{1/2}}{3\pi}. \quad (\text{A2.2})$$

Using the standard definition of the Fermi energy

$$N_\tau = \int_0^{\epsilon_f} g_\tau(\epsilon) d\epsilon = \frac{4}{9\pi} \left(\frac{2\mu\epsilon_f R_0^2}{\hbar^2} \right)^{3/2}, \quad (\text{A2.3})$$

we get for the single-particle state density at the Fermi surface the formula

$$g_\tau(\epsilon_f) = \left(\frac{2}{3\pi^2} \right)^{1/3} \frac{2\mu r_0^2}{\hbar^2} N_\tau^{1/3} A^{2/3}, \quad (\text{A2.4})$$

that differs from Eq. (8) by the numerical coefficient only.

The well-known formula for the single-particle state density of a two-component Fermi gas [22] can be derived from Eq. (A2.4) in case of $Z=N=A/2$:

$$g(\epsilon_f) = g_Z(\epsilon_f) + g_N(\epsilon_f) = \frac{4\mu r_0^2}{(3\pi^2)^{1/3} \hbar^2} A \quad (\text{A2.5}).$$

Eq. (7), that is widely used in various applications, is based on a simplified linear splitting of Eq. (A2.5).

The difference between the numerical coefficients in Eqs. (7), (8) and (A2.4), (A2.5) reflects a possible contribution of collective excitations and other effects not included in the semi-classical approach.

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