Excitation-energy sorting in pre-scission dynamics

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Abstract: The thermodynamically driven processes in pre-scission dynamics are investigated. During the shape evolution towards scission, the two nascent fragments develop their individual properties. The fact that the different temperatures of the two fragments do not depend on excitation energy, which is deduced from recently measured level densities, leads to an excitation-energy sorting process, where all intrinsic excitation energy is transferred to the cold fragment. This process has an impact on the emission of prompt neutrons and gamma rays from the two fragments. If the energy-sorting process is completed, before the exchange of protons through the neck becomes inhibited, the hotter fragment is formed with a preferentially even number of protons.

Calculations with a schematic model demonstrate that the proposed scenario accounts for several complex features of prompt neutron emission and of the even-odd effect in fission-fragment yields, which remained unexplained up to now. The importance of nuclear fission as a laboratory for studying the dynamics of non-equilibrium processes between mesoscopic superfluid objects is stressed.

Introduction

Most objects in nature have an approximately constant number of degrees of freedom, and their temperature, defined as the average excitation energy per degree of freedom, increases with increasing total excitation energy E^* of the system. However, nuclei with moderate E^* behave very differently. Experiments on nuclear level densities have shown that at least up to E^* = 6-7 MeV the temperature of nuclei does essentially not change with increasing E^* [1]. Moreover, it was even found recently that for medium-mass nuclei the temperature stays constant up to E^*= 20 MeV [2]. The main reason for this constant-temperature behaviour is that pairing correlations lead to an effective number of degrees of freedom that increases in proportion to E^*. Cooper pairs of neutrons and protons melt in a way that the mean energy per nucleonic excitation and thus the nuclear temperature stays constant. In nature, this behaviour appears in first-order phase transitions (e.g. solid-liquid or liquid-gas). In a mixture of two phases, like ice and water, the temperature of the mixture remains constant when energy is introduced or extracted, as long as both phases are present. Only the fractions of the two phases vary. It is of special interest to study, how two mesoscopic quantum-mechanical objects in such a particular regime of constant temperature behave when they are in thermal contact. The scission configuration in the nuclear-fission process, where two different nuclei can exchange E^* through the neck, offers a unique possibility to investigate this phenomenon.

Energy balance at scission

In fission, the energy difference between the ground-state masses of the initial fissioning system and the final fission fragments, given by the Q value, and the initial excitation energy of the fissioning nucleus E^*_{CN}, end up either in the total excitation energy (*TXE*) or in the total kinetic energy (*TKE*) of the fragments. The *TXE* is available for particle evaporation and gamma emission either before scission or from the separated fragments. In this work, we consider low-energy fission with initial excitation energies E^*_{CN} up to a few MeV where evaporation and gamma emission on the fission path is considered to be weak. The same is true for neck emission of neutrons. Since fission fragments are neutron-rich, evaporation proceeds almost exclusively by neutrons. We assume that already somewhat before the scission configuration the two nascent fragments have acquired their individual properties concerning shell effects [3,4,5] and pairing correlations [6] and can be treated as two well defined nuclei set in thermal contact through the neck. Theoretical investigations of the gradual transition from the mononucleus regime to the di-nuclear system [3,4,5,6,7,8] support this assumption.

We will now consider how the *TXE* is divided between the two nascent fragments. Following the transition-state approach of Bohr and Wheeler [9], all the available *E** above the barrier height is assumed to be thermalised, that means it is, on the average, equally distributed between all available intrinsic and collective degrees of freedom. These are the single-particle excitations and the collective normal modes. On the way to scission, the difference in potential energy between saddle and scission [10] may feed some amount of pre-scission kinetic energy in fission direction, excitations of normal collective modes and additional intrinsic excitations.

We may distinguish three classes of energy, which add up to the final *TXE* of the fission fragments, according to their appearance at scission: (i) Collective excitations stored in normal modes. (ii) Intrinsic excitations by single-particle or quasi-particle excitations. (iii) Deformation energy. The deformation energy ends up as part of the *E** available when the fission fragments recover their ground-state deformations.

The deformation induced in the two nascent fragments can be considered as a superposition of a macroscopic trend, caused by the mutual Coulomb repulsion of the nascent fragments, which favours a large prolate deformation around $\beta = 0.5$ [11] and a structural influence due to shell effects. Different fission modes correspond to substantially different deformations at scission and, thus, to different amounts of deformation energy of the individual fragments. Theoretical arguments on the deformation of the fragments at scission can be deduced from shell-model calculations [11,12], while experimental information can be extracted from the saw-tooth-like behaviour of the neutron yields, which is thought to be caused to a great extent by the variation of the contribution of the deformation energy to the *E** of the fragments.

The division of collective excitations among the two fragments is intimately related to the nature of the specific collective mode considered. As an example, the division of *E** stored in angular-momentum-bearing modes is governed by the momenta of inertia of the fragments and the conservation of total angular momentum. If the fissioning nucleus has zero angular momentum, and orbital angular momentum is neglected, both fragments must carry the same amount of angular momentum (in opposite direction), and, thus, the *E** is inversely proportional to their moment of inertia. Thus, for these specific modes, the lighter fragment tends to carry the larger portion of *E**.

Division of intrinsic excitation energy

The division of intrinsic excitations can be derived when thermal equilibrium at scission is assumed among the intrinsic degrees of freedom in each fragment. As said above, the nuclear level density at low *E** is very well described by the constant-temperature formula:

$$\rho\,(E^*) \propto \exp(E^*/T) \qquad (1)$$

In a recent work, Egidy et al. have obtained the following dependence of the nuclear temperature *T* with the nucleus mass number *A* and with shell effects *U* from a fit to available data on nuclear level densities [13] :

$$T = \frac{1}{A^{2/3}}(17.45 - 0.51\,U + 0.051\,U^2) \qquad (2)$$

This leads to a very interesting situation for the two nascent fragments at the scission-point configuration: The level density of each fragment is represented by the constant-temperature formula (1) with a specific value of *T* for each fragment. As a consequence, there is no solution for the division of intrinsic *E** with $T_1 = T_2$. As long as some excitation energy remains in the fragment with the higher temperature, its *E** is transferred to the fragment with the lower temperature. That means, a process of *E** sorting takes place where all *E** accumulates in the fragment with the lower temperature, while the other fragment looses its entire *E**. According to formula (2) the heavy fragment generally has the lower *T* and thus attracts all the *E**. Some deviations from the constant-temperature behaviour appear in the range of the first quasi particle excitations [14].

Due to the influence of shell corrections on *T*, see eq. (2), the direction of the energy transfer may be reversed if the heavy fragment is stabilised by a strong shell effect. This may be possible in the standard I (SI) fission channel, which is characterised by the formation of a heavy fragment close to the doubly magic ^{132}Sn.

The flow of excitation energy from the hot fragment to the cold fragment is a way for the entire system made of the two nascent fragments in contact to maximise the number of occupied states or its entropy. In fact, the entropy S is a linear function of the partitioning of the total excitation energy $E^* = E^*_1 + E^*_2$:

$$S = S_1 + S_2 = \frac{E_1^*}{T_1} + \frac{E_2^*}{T_2} = \frac{E_1^*}{T_1} + \frac{E^* - E_1^*}{T_2} = \frac{E^* T_1 + (T_2 - T_1) \cdot E_1^*}{T_1 \cdot T_2} \qquad (3)$$

The number of available states of the light nucleus or closed-shell nucleus is small compared to that of the complementary fragment. Therefore, the situation in which the light nucleus or the closed-shell nucleus has part of the E^* leads to a smaller entropy than the situation in which the entire E^* is transferred to the heavy or the non-closed-shell nucleus which has considerable more available states.

Prompt neutron yields

The number of evaporated neutrons as a function of the fragment mass is directly related to the excitation energy of the fragment and, therefore, should clearly reflect the peculiar situation of the full transfer of the intrinsic excitation energy to the cold fragment. The neutron-induced fission of ^{237}Np has been studied very carefully at two different neutron energies [15]. Fig. 1 shows the average number of evaporated neutrons as a function of the fragment mass. As mentioned above, the well known saw-tooth-like behaviour of this curve is attributed to the deformation energy. The minimum close to A=130 is due to the shell closures N=82, Z=50 that lead to spherical fission fragments. An increase of incident neutron energy translates into an increase of E^* of the compound nucleus. The increase of the emitted neutrons near symmetry for 110<A<130 with incident neutron energy is caused by the increase of the yield of the super long (SL) mode which is related to well deformed fission fragments. For more asymmetric mass splits outside this range, we observe a very peculiar feature: Interestingly, Fig. 1 shows that the increase of E^* leads to an increase of the number of evaporated neutrons for the heavy fragment, only. Since the neutron yield of the fission fragments for a fixed mass fluctuates over several neutrons, the mean value is a very sensitive measure of the fragment excitation energy. If the mean energy available changes, the contribution on one or the other wing of the neutron-multiplicity distribution decreases respectively increases, and, thus, the mean value is shifted. Actually, a quantitative analysis of the data reveals that all of the increased E^* appears in the heavy fragment. This observation is rather general as it was also found for other fissioning systems such as ^{233}U and ^{238}U and other incident particles like protons [16,17,18,19]. However, no clear explanation has yet been found for this effect. The reason is that all the work [20,21,22] done to study the partition of intrinsic excitation energy between fission fragments is based on the formula of Bethe [23].

$$\rho(E^*) \propto \exp(2\sqrt{a E^*}) \qquad (4)$$

where a is the level-density parameter which is proportional to the mass number A of the nucleus. The latter formula is based on independent particles in an equidistant single-particle level scheme. Under the assumption of thermal equilibrium at scission, one obtains an intrinsic E^* division in proportion of the mass ratio of the fragments: $E^*_1 / E^*_2 = A_1 / A_2$.

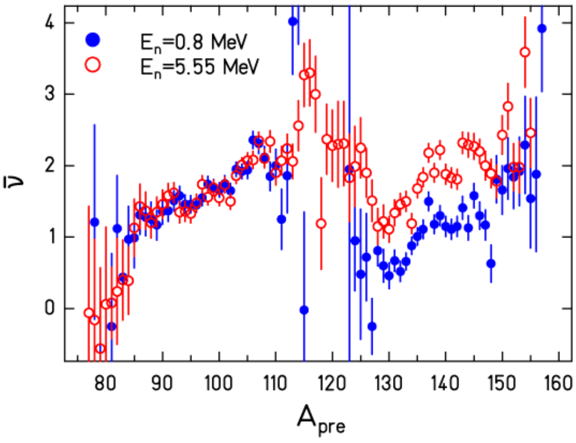

Figure 1. *Average number of prompt neutrons as a function of the primary fragment mass for the neutron-induced fission of* ^{237}*Np at two incident-neutron energies, data taken from ref. [15].*

The tendency to divide the available excitation energy according to the mass ratio has been confirmed empirically in many binary reactions involving relatively high E^* [24], although also deviations from full equilibration were observed due to insufficient reaction time [25]. However, this expression is not applicable at low E^* and fails to explain the observation presented in Fig. 1 that all the increase in E^* is found in the heavy fragment. Actually, this effect is a direct consequence of the different constant temperatures of the two fragments at scission. According to eq. (2), the temperature of the heavy fragment, in the absence of strong shell effects, is always lower than the temperature of the light fragment. Therefore, the heavy fragment will absorb the entire available intrinsic E^* and evaporate more neutrons.

We would like to stress that our argumentation is based on the same assumptions as other work that investigates the sharing of intrinsic E^* at scission [20,21,22]. That is, we have assumed independent fission fragments and a process of thermal equilibration between the fragments at scission. What is substantially different in our approach is that we use the constant-temperature level density which correctly describes the behaviour of nuclei at moderate E^* and not the commonly used Fermi gas level density of eq. (4) which is only valid at high E^*.

Features of the even-odd effect in fission

Pairing correlations are not only at the origin of the constant-temperature behaviour of the nuclear level density, they manifest themselves also in a number of observables, which are modulated by an even-odd structure [26]. 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 2 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) [27]. 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.

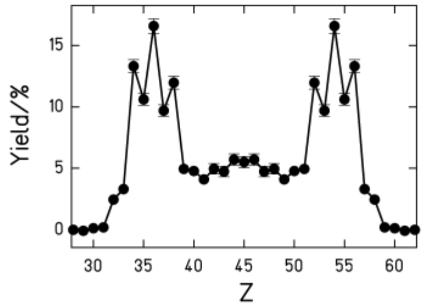

Figure 2. *Element distribution observed in the electromagnetic-induced fission of ^{229}Th [28].*

At present, several systematic features have been established experimentally [29]. The left part of Fig. 3 shows experimental data on the local even-odd effect δ_p, defined as the logarithmic four-point difference [30] 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 (refs. [28,31,32]) and from figure 3 clearly illustrate several features:

(i) The amplitude δ_p 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.

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 [26,31]. 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 [33]. It seems 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 characterised by low yields and a higher barrier in the heavier actinides [34].

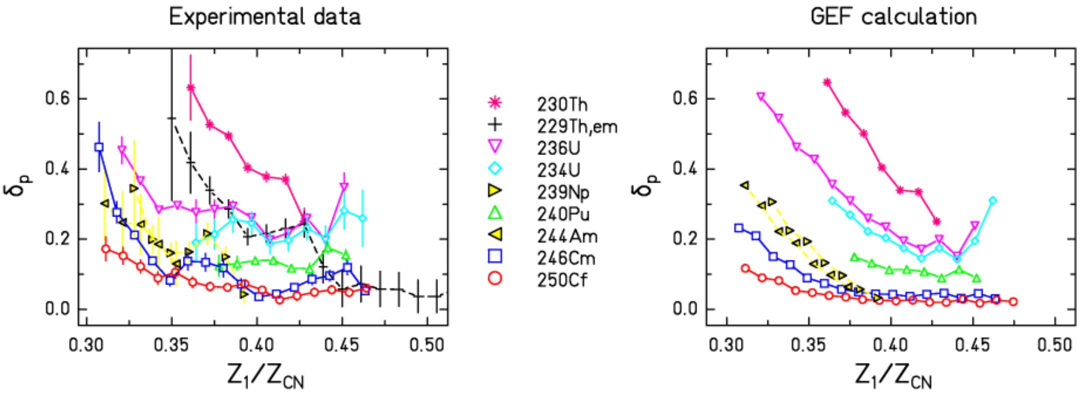

Figure 3. *Left part: Systematics of the measured local even-odd effect as a function of asymmetry, parameterised as the ratio of the Z of the light fragment and the Z of the fissioning nucleus Z_1/Z_{CN}, for thermal-neutron-induced fission of heavy nuclei. The fissioning nucleus is indicated. The data have been taken from ref. [29]. The local even-odd effect of the electromagnetic-induced fission of ^{229}Th [28] (see figure 2) is shown in addition. Right part: Result of a calculation with the GEF code [35].*

Some attempts were made to theoretically study the dynamical process of pair breaking in the fission process [6,31,36,37]. 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 [38] or the single-particle level density [28,39], but they stayed on a purely statistical level, and the quantitative agreement with the data was not satisfactory.

Even-odd effect in asymmetric fission and energy sorting

In the present work, we apply our considerations on the energy-sorting mechanism in superfluid fission dynamics [40] to propose a dynamical scenario for the asymmetry-associated even-odd effect in fission. Since the temperature of the fragments remains unchanged in spite of the variation of E^*, the light fragment will transfer all its E^* to the heavy one. 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, there will be a tendency for the hot (normally the light) fragment to be fully paired.

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 all 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 all the E^*. The latter will increase with the initial excitation energy in the light fragment $E^*_{0,\text{light}}$ since it will take a longer time to transfer all the energy from the light to the heavy nucleus. We consider that the initial excitation energy $E^*_{0,\text{light}}$ 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 $E^*_{sad-sci}$ increases with the Coulomb parameter $Z^2/A^{1/3}$ since the saddle-to-scission path becomes longer [10]. 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. To resume, the time t follows the expression:

$$t \propto \frac{E^*_{sci}}{T_1 - T_2} \tag{5}$$

As a consequence, t will increase with the beam energy and the Coulomb parameter of the fissioning nucleus and will decrease with increasing asymmetry of the mass split. Eq. (5) is reflected by the schematic drawing shown in Fig. 4, 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.

Let us now assume that 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 $t > t_p$, no net even-odd effect is induced because protons cannot be transferred to the heavy nucleus. 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. 4, 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. 3 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.

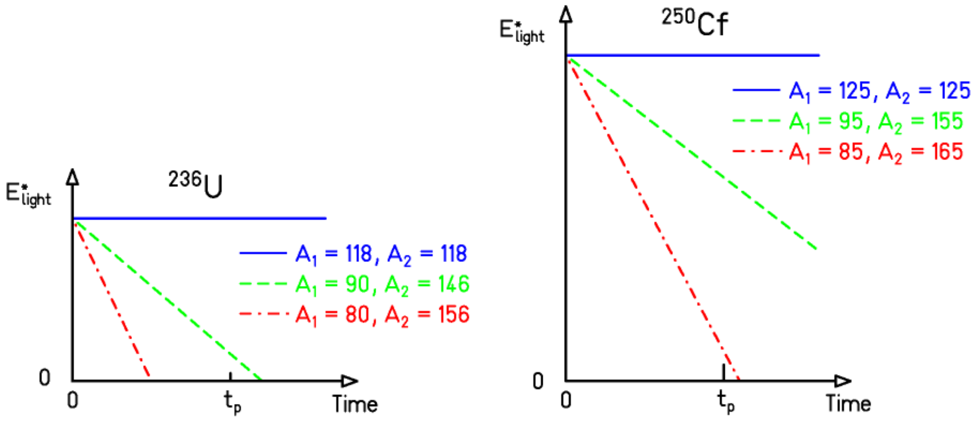

Figure 4. *Schematic drawing representing the mean excitation energy in the light fission fragment as a function of time. Two fissioning nuclei and different mass splits are considered. See text for details.*

For several systems, the data point in Fig. 3 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 deduced even-odd effect. In the GEF code [35] the dependence of the local even-odd effect with asymmetry is modelled in a phenomenological way with a smoothed step function, obtained by a convolution with a Gaussian function. The threshold asymmetry value 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. In accordance with the data, the width of the Gauss 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 the GEF model it is assumed that 50% of the energy release from saddle to scission [10] is dissipated into intrinsic excitations. The intrinsic excitation energy at scission determines also the magnitude of the even-odd effect at symmetry according to the model of ref. [41].

On the right part of Fig. 3, the results of the GEF code 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, one expects smaller threshold asymmetries for the even-odd effect in the fission-fragment neutron yields. Unfortunately there are no data to verify these statements.

Summary and outlook

Nuclei at low excitation energy E^* are peculiar systems, since their temperature remains approximately constant with increasing E^*. In this sense, the nuclear superfluid to normal-liquid phase transition seems to behave like a first-order phase transition. The very special feature of this phenomenon in nuclei is that the constant-temperature regime essentially reaches down to zero energy, with only some fluctuations at the thresholds for the first quasi-particle excitations. The scission configuration of the fission process offers the unique possibility to investigate, how two different nuclei in this special regime of constant temperature share the available intrinsic excitation when they are in thermal contact. We have shown that in this regime we reach a peculiar state of thermal equilibrium at scission in which the temperatures of the nascent fragments remain different in spite of the flow of E^* from the hot to the cold fragment. Rather unexpectedly, this implies that the total amount of intrinsic E^* available at scission is found in the fragment with the lower temperature. Our discovery of the energy-sorting mechanism may be considered as a new counter-intuitive manifestation of quantum-mechanical properties of microscopic systems. This entropy-driven E^*-sorting process appears to have similarities with Maxwell's demon [42] on the nucleonic level. However, the phenomenon is fully compatible with the second law of thermodynamics. This E^*-sorting effect explains very easily an issue that remained unsolved up to present when comparing the number of emitted neutrons as a function of fragment mass for different initial excitation energies. It was observed in asymmetric mass splits that the increase of intrinsic E^* of the fissioning nucleus appears as an increase of E^* in the heavy fission fragments, only.

Moreover, 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, generally from the light to the heavy fragment, at the last step of the energy-sorting process. The fact that the even-odd effect is governed by the ratio of the total intrinsic excitation energy at scission and by the temperature difference of the two nascent fragments lead us to propose a schematic dynamical model.

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 [43], but there is little knowledge on the time for intrinsic excitation-energy transfer between nuclei in thermal contact in the superfluid regime. Thus, the present work is a step forward in the development of new kinds of fast nuclear clocks. Detailed theoretical and experimental studies on pre-scission dynamics will allow extending the investigations on non-equilibrium processes between different superfluid mesoscopic objects in analogy to the supercurrent [44] 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 findings provide an important constraint on the theoretical modelling of the last stage of fission in the superfluid regime [6], which represents still a considerable challenge.

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