Review

Review on the progress in nuclear fission—experimental methods and theoretical descriptions

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Abstract
An overview is given on some of the main advances in the experimental methods, experimental results, theoretical models and ideas of the last few years in the field of nuclear fission. New approaches have considerably extended the availability of fissioning systems for the experimental study of nuclear fission, and have provided a full identification of all fission products in A and Z for the first time. In particular, the transition from symmetric to asymmetric fission around 226Th, some unexpected structures in the mass distributions in the fission of systems around Z = 80–84, and an extended systematics of the odd–even effect in the fission fragment Z distributions have all been measured (Andreyev et al 2018 Rep. Prog. Phys. 81 016301). Three classes of model descriptions of fission presently appear to be the most promising or the most successful. Self-consistent quantum-mechanical models fully consider the quantum-mechanical features of the fission process. Intense efforts are presently being made to develop suitable theoretical tools (Schunck and Robledo 2016 Rep. Prog. Phys. 79 116301) for modeling the non-equilibrium, large-amplitude collective motion leading to fission. Stochastic models provide a fully developed technical framework. The main features of the fission-fragment mass distribution have been well reproduced from mercury to fermium and beyond (Möller and Randrup 2015 Phys. Rev. C 91 044316). However, limited computer resources still impose restrictions, for example, on the number of collective coordinates and on an elaborate description of the fission dynamics. In an alternative semi-empirical approach (Schmidt et al 2016 Nucl. Data Sheets 131 107), considerable progress in describing the fission observables has been achieved by combining several theoretical ideas, which are essentially well known. This approach exploits (i) the topological properties of a continuous function in multidimensional space, (ii) the separability of the influence of fragment shells and the macroscopic properties of the compound nucleus, (iii) the properties of a quantum oscillator coupled to a heat bath of other nuclear degrees of freedom, (iv) an early freeze-out of collective motion, and (v) the application of statistical mechanics for describing the thermalization of intrinsic excitations in the nascent fragments. This new approach reveals a high degree of regularity and allows the calculation of high-quality data that is relevant to nuclear technology without specifically adjusting the empirical data of individual systems.
Keywords: nuclear fission, innovative experiments, microscopic and global models, topographic theorem, early influence of fragment shells, entropy-driven energy sorting, hidden regularity in fission observables

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1. Introduction

The discovery of nuclear fission revealed that the heaviest nuclei are barely bound in their ground state. An excitation energy on the order of a few per thousand of their total binding energy is sufficient to induce disintegration into two large pieces, releasing a huge amount of energy of about 200 MeV. Thus, the specific energy content of nuclear fuel is about $10^8$ times larger compared to fossil fuels like coal, mineral oil or natural gas. This explains the importance of fission in nuclear technology.

The energy stored in heavy nuclei, and even the synthesis of an appreciable portion of matter in the Universe, has its origin in the astrophysical r-process, a process of consecutive neutron capture and beta decay in an environment with a very high neutron flux at some astrophysical site, which has not fully been identified for long [1–3]. The recent observation of the merging of a binary neutron-star system [4] affirmed the importance of this scenario for r-process nucleosynthesis.

Fission is believed to play an important role in the r-process itself by fission cycling, which limits the mass range of the r-process path and has an influence on the associated nuclide abundances [5, 6]. The r-process nucleosynthesis cannot be fully understood without a good knowledge of the fission properties of the very neutron-rich isotopes of the heaviest elements, which are presently not accessible to direct measurements [7, 8]. Therefore, additional interest for a better
understanding of the fission properties of nuclei far from stability comes from astrophysics.

In a general sense, nuclear fission offers a rich laboratory for a broad variety of scientific research on nuclear properties and general physics. The relatively flat potential energy of fissionable nuclei reaching very large deformations, when compared to lighter nuclei, allows the study of nuclear properties like shell effects in super- and hyper-deformed shapes [9]. Phenomena connected with the decay of the quasi-bound nuclear system beyond the fission barrier yield information on nuclear transport properties like nuclear viscosity [10, 11] and heat transfer between the nascent fragments [12]. They even offer a valuable test ground of general importance for non-equilibrium processes in isolated mesoscopic systems, where quantum mechanics and statistical mechanics play an important role [13].

During the last few years, there has been considerable activity in the field of nuclear fission, both experimental and theoretical. Several detailed and some comprehensive papers have been written on the development of theoretical approaches and formalisms with an impetus on fission dynamics and its signatures. This is illustrated by a representative list of papers that have been published in the last seven years [11, 14–98]. The description of fission cross sections and fission probabilities in induced fission, which involves the entrance channel, transmission through the fission barrier and competition with other exit channels, is a field of continuous activity [99–109]. In this domain, the coupled-channel model, the dispersive optical potential, and a heuristic model for resonant transmission are of eminent importance. On the experimental side, there have been several publications on the refinement of existing techniques [110–115] or the development of novel ones [116, 117] as well as on new experimental findings [118–196]. Usually, the technical development of the specific theoretical or experimental approach, its challenges and achievements, are in the focus of these papers. Theoretical papers often intend to demonstrate the quality of a specific approach by showing its ability to reproduce some distinct data and to present computational algorithms that provide suitable approximate descriptions when exact solutions are out of reach, which is often the case.

Experiments and evaluation\(^1\) are often driven by the interest in the reliable nuclear data required for technical applications, mostly in reactor technology. In this context, we mention for example (i) the measurements of prompt-neutron spectra at the LANSCE accelerator facility at the Neutron Science Center in Los Alamos, New Mexico, USA [198], (ii) fission-fragment yield, cross-section, prompt-neutron and gamma-emission data from actinide isotopes at the Joint Research Centre in Geel (previously called IRMM), Belgium [199], (iii) a coordinated research project (CRP) on the evaluation of the prompt-fission-neutron spectra of actinides, organized by the IAEA, Vienna, Austria [200], (iv) measurements of the energy dependence of fission-product yields from \(^{235,238}\text{U}\) and \(^{239}\text{Pu}\) at TUML, Durham, North Carolina, USA [201], (v) MCNP studies on multiple scattering contributions, over-corrected backgrounds, and inconsistent deconvolution methods used in the evaluated prompt-fission-neutron spectra of \(^{239}\text{Pu}(n,f)\) [202], (vi) the high-precision measurement of the prompt-neutron spectrum of \(^{252}\text{Cf}\) at the Rensselaer Polytechnic Institute, Troy, New York, USA [203], (vii) a measurement of the energy and multiplicity distributions of neutrons from the photo-fission of \(^{235}\text{U}\), again at Los Alamos Neutron Science Center [204], and (viii) measurements of the beta decay of fission products with the modular total-absorption spectrometer at the Holifield Radioactive Ion Beam Facility, Oak Ridge, Tennessee, USA [205, 206]. We would also like to mention that there has been a European initiative in the last few years to measure the neutron-induced fission cross section of \(^{242}\text{Pu}\), using very different experimental methods in order to provide several completely independent measurements of this cross section. This is the only means of strongly reducing the final systematic uncertainties in this cross section to comply with the very demanding uncertainty required by reactor applications [207]. Measurements of the \(^{242}\text{Pu}(n,f)\) cross section were recently carried out, for example, at the JRC Geel [177], at the National Physical Laboratory in Teddington, UK [208], at the Van de Graaff accelerator of Bruyères le Châtel, France [209], at the Physikalisch-Technische Bundesanstalt in Braunschweig, Germany [210] and at the nELBE facility of the Helmholtz-Zentrum Dresden—Rossendorf, Germany [211].

The present review article has a different goal: it aims to promote an improved understanding of the nuclear-fission process by establishing a synopsis of different theoretical approaches and of empirical knowledge on a general level. Its impetus lies in tracing back experimental findings to the underlying physics on different levels, reaching from microscopic descriptions to statistical mechanics while covering essentially all fission quantities.

We carefully consider the fission barrier, because it is a key quantity for establishing the probability of fission occurring in competition with other decay channels. In particular, it is important for determining the individual contributions when modeling multi-chance fission, which is fission after the emission of one or more neutrons. While direct information on the evolution of the system between saddle and scission is scarce, the fission-fragment yields, the total kinetic energies and the prompt-neutron multiplicities are the most important observables that result from this evolution by evidencing the division of the protons and neutrons as well as the magnitude and division of the excitation energy between the fragments. The energy spectra of prompt neutrons, [200] and the references therein, and prompt gammas [128, 137, 139, 146, 148, 152, 161, 186] are calculable in a rather straightforward manner within the statistical model, once the conditions at scission (fission-fragment distributions in mass and atomic number, excitation energy and angular momentum) are imposed. This is demonstrated by the rather successful attempts [213–227],
in cases where the fragment properties mentioned above are empirically known or can be estimated. The latter work is motivated by the interest of nuclear technology to better reproduce the measured energy spectra in order to control the irradiation load and the heat production in fission. The GEF model [228] (see section 4.3) also gives a rather accurate reproduction of the measured spectra and other characteristics of the prompt neutrons and prompt gammas. In this case, the conditions at scission are provided by the model itself. Therefore, the de-excitation process does not carry so much information on the fission process, if we disregard the quest for scission neutrons, which are neutrons of non-statistical nature emitted at scission [200], and whose existence has been controversially discussed [229], as well as efforts to better comprehend the generation of angular momentum of the fission fragments, which is still not well understood [13].

We will concentrate on low- and medium-energy fission, where binary fission, ending up in two heavy fragments, is the dominant decay channel. Ternary fission with its very specific features is not included. A compact but rather exhaustive record of the most relevant experimental and theoretical work in the field of ternary fission is given in the introduction [230]. The decay of highly excited nuclei, where the phase space favors multi-fragmentation [231, 232], and which involves simultaneous decay into more than two fragments, and quasi-fission [233], after heavy-ion reactions which preserve a memory on the entrance channel, are not covered either.

The present article is structured as follows. After a short reminder of the former status of knowledge in section 2, we will give a review on recent innovations in the experimental and theoretical work in sections 3 and 4, respectively. In detail, major steps in experimental fission research which have been made in the last few years by the application of inverse kinematics and by technical innovations in the application of beta-delayed fission will be reported. On the theoretical side, the applications and further developments of fully dynamical descriptions of the fission process in quantum-mechanical and classical models will be presented in sections 4.1 and 4.2. In addition, the application of a number of general laws, concepts and theorems in a semi-empirical model, described in sections 4.3 and 4.4, has delivered very interesting explanations or opened well-targeted questions for some prominent and some very peculiar observations that had remained unexplained for a long time, or that emerged from the results of recent experiments. Models on the specific aspects of fission are presented in section 4.5. A general discussion of current problems, which covers the experimental results and different theoretical models and ideas is provided in section 5, followed by a discussion on the expected evolution of fission theory and experiments to satisfy the main identified needs. A full scenario for fission is then proposed, and the main associated theoretical achievements are highlighted. Finally, a summary is found in section 6.

In the interest of the comprehensive and consistent discussion in section 5, section 2 does not include former experimental results that have only been interpreted recently in the framework of new theoretical ideas. In a similar way, section 4 predominantly presents the basic and technical aspects of the different models.

2. Former status of knowledge

Since the discovery of nuclear fission [234, 235], the bulk of experimental results has been obtained in the neutron-induced fission of available and manageable target nuclei. Limitations arose from the small number of primordial or long-lived heavy target nuclides and from the technical difficulties of experiments with mono-energetic neutrons of arbitrary energy. Therefore, as well as for some other practical and technological reasons, the majority of experiments on fission yields have been made with thermalized reactor neutrons and, to a lesser extent, with fast neutrons (which denote typical energies of unmoderated or slightly moderated fission neutrons) and neutrons of 14 MeV, e.g. from the $^3$H + $^1$H $\rightarrow$ n + $^4$He reaction. Moreover, in all experiments performed in direct kinematics, the kinetic energies of the fission products are hardly sufficient for obtaining the unambiguous identification of fission products in A and Z in kinematical measurements, which implies that complete fission-product distributions on the N-Z plane cannot be obtained.

An exhaustive overview of the experimental results on neutron-induced fission has been given in a recent review by Gönnenwein [236]. It essentially covers fission cross sections, the fission-product mass distributions, kinetic and excitation energies, nuclear-charge distributions in the group of lighter fission products with an emphasis on the odd–even staggering, and the emission of prompt neutrons and gammas.

Also, many experiments on charged-particle-induced fission [237] and photon-induced fission (bremsstrahlung or mono-energetic photons), e.g. [163] and the references therein, have been and are still being performed. The use of charged-particle or heavy-ion projectiles made a number of additional nuclei available for fission experiments by transfer reactions [238] or fusion (e.g. [239–241]). The easy accessibility of higher excitation energies and the inevitable population of larger angular momenta has allowed other aspects of the fission process to be studied. They are described in the recent review by Kailas and Mahata [237], and are not covered in this work.

The prominent theories developed in parallel with these observations have provided the potential-energy surfaces of fissioning systems in macroscopic–microscopic [242] or in self-consistent microscopic approaches [243]. Brosa et al. [244] associated gross structures, which are manifested experimentally by clear structures in the mass and nuclear-charge yields, prompt neutron yields and fragment total kinetic energies (TKE) with valleys in the multi-dimensional potential, which imply particularly stable combinations of mass splits. The nomenclature introduced in [244], which distinguishes the ‘super-long’ (SL) symmetric fission mode and the asymmetric ‘standard’ fission modes in sequence of increasing mass asymmetry (S1, S2), is still used. Often, they are labeled as fission channels. Fine structures in the fission-fragment mass and nuclear-charge distributions were analyzed with
the combinatorial model of Nifenecker et al [245]. However, both models [244, 245] have rather remained on the level of empirical parametrizations. After the first qualitative considerations on the dynamical evolution of the fissioning system at low excitation energies, e.g. by the S-matrix formulation of Nörenberg [246], two types of microscopic quantum-dynamic calculations were performed quite early on. By 1978, the time-dependent Hartree–Fock method [247] had already been applied to fission. Later, time-dependent calculations based on the generator-coordinate method using Hartree–Fock–Bogoliubov states were performed, and the most probable fission configuration of $^{240}$Pu was analyzed [243]. Wilkins et al [248] performed quantitative calculations of fission quantities, for example fission-fragment mass distributions, charge polarization (which leads to different $N/Z$ ratios of the two complementary fragments), total kinetic energies and prompt-neutron multiplicities, with a static statistical scission-point model, including the influence of shell effects and pairing correlations. Although this model disregards any influence of the dynamics, which prevents, for example, any information on dissipation or fission times being obtained, successors of this model are still being developed [37, 50, 53]—often achieving a good reproduction of measured mass distributions and other quantities, for example for the thermal-neutron-induced fission of $^{232}$Th, $^{235}$U, $^{239}$Pu and $^{241}$Cm [37], and for the spontaneous fission of nuclei around $^{250}$Fm and above [50].

The status of experimental and theoretical research described in this section, which we denote as former knowledge, was achieved around the 50th anniversary of the discovery of fission in 1989, when the ‘conventional’ technical possibilities of experimental equipment and computer power, respectively, had been exploited to a large extent. Considerable progress has been achieved in the following years up to the present day, mostly due to novel experimental methods and the exponentially developing possibilities for performing appreciably more complex theoretical calculations, as described in the following sections.

3. Experimental innovations

There has been continuous progress in the quality of experimental equipment through the development of technology in many fields. This has allowed us to improve the quality and to extend the quantity of experimental results in many aspects. As a direct consequence, the data basis for applications in nuclear technology has improved considerably. In the present section, we give a concise overview of only a few of the major developments that have given us a considerably improved insight into the physics of the fission process. A comprehensive and detailed overview of technological developments and new experimental results is presented in a dedicated review, which appeared very recently in the same journal [249].

$^2$The charge polarization is defined as $\Delta Z = \langle Z \rangle - Z_{UCD}$ with $Z_{UCD} = A \cdot Z_{CN}/A_{CN}$, $\langle Z \rangle$ is the mean nuclear charge for a given fragment mass $A$, and CN denotes the fissioning nucleus.

$^3$See section 4 for detailed information about the different theoretical approaches.

3.1. Accessible fissionable nuclei

The progress in understanding fission has heavily relied on, and still relies on, the development of advanced experimental methods. A severe restriction remains the availability of fissionable nuclei as a target material. Therefore, the traditional use of neutrons for inducing fission offers only a rather limited choice of fissioning systems. These limitations are increasingly being overcome by alternative methods: for instance, spontaneously fissioning heavy nuclei have been produced by fusion reactions [250] for many years, as already mentioned. These experiments provide fission-fragment mass distributions for very heavy nuclei, finally limited by statistical uncertainties due to the low number of nuclei being produced [241].

Recently, very neutron-deficient nuclei, e.g. in the $Z = 80$ region, were produced in spallation reactions at ISOLDE [251], CERN, which undergo beta-delayed fission [252]. This experiment profited from an unambiguous identification of the fissioning nuclei, mass selection by ISOLDE, and Z selection by the resonance-ionization laser ion source RILIS [253]. A pronounced double-humped mass distribution was found for the fission fragments of the compound nucleus $^{180}$Hg, formed as the daughter nucleus after the beta decay of $^{180}$Tl, which has similarities with the double-humped mass distribution observed previously by Itkis et al [254] for the fission of excited $^{201}$Tl, which is situated close to beta stability, in an alpha-induced reaction. Fission-fragment mass distributions have also been studied in the beta-delayed fission of the daughter nuclei $^{184}$Hg and $^{194,196}$Po at ISOLDE and $^{202}$Rn at SHIP (GSI) [252], where asymmetric fission or complex shapes were observed. These unexpected observations triggered several experiments with different techniques (heavy-ion fusion–fission reactions and electromagnetic-induced fission in inverse kinematics) and a number of theoretical works with self-consistent and stochastic models, which will be described in detail in the following. Again, by the study of beta-delayed fission at ISOLDE, Ghys et al [254] found single-humped fission-fragment mass distributions in the beta-delayed fission of the daughter nucleus $^{194}$Po and indications for triple-humped distributions for $^{196}$Po and $^{202}$Rn. Asymmetric fission was observed following the population of $^{182}$Hg at an excitation energy of 22.8 MeV above the saddle point, while symmetric fission was observed for $^{195}$Hg nuclei at a similar excitation energy above the saddle point in heavy-ion fusion–fission reactions [175]. In this case, the situation is complicated by the possibility of multi-chance fission, which means contributions from the fission of neighboring nuclei at lower excitation energy after the pre-fission emission of neutrons and, in the case of very neutron-deficient systems, also protons. Multi-chance fission should also be considered when interpreting the measured mass distributions of $^{180}$Hg and $^{190}$Hg, formed in $^{36}$Ar-induced reactions [255], where asymmetric fission was found, and for the measured mass distributions of $^{179}$Au and $^{189}$Au, formed in $^{35}$Cl-induced reactions [256], where symmetric fission was observed for the lighter system, while the heavier system showed indications for an asymmetric component. These experiments have revealed the complexity of fission-fragment mass distributions in the lead
region, but they are still too fragmentary to establish a full systematics of variations with the fissioning system. Although beta-delayed fission is especially suited for studying low-energy fission, it is very restricted to nuclei with beta Q values above or not much below the fission threshold as well as high fission probabilities. These are mostly odd–odd very neutron-deficient nuclei. Alternative techniques are required to establish a more complete systematics of low-energy fission in the lead region. The most promising one is the electromagnetic fission of secondary beams at relativistic energies, which will be mentioned below.

Advanced experimental studies on the light-charged-particle-induced fission probabilities of systems that are not accessible by neutron-induced fission are being performed systematically. These surrogate-reaction studies focus on the ability of these alternatives to simulate neutron-induced reactions [257]. Also recently, the use of heavier ions, for example $^{16}$O, in transfer reactions has allowed us to appreciably extend the range of fissionable nuclei available for fission studies and to measure the fission-fragment mass distributions over an extended range of excitation energy [191] at the JAEA tandem facility at Tokai. Moreover, comprehensive studies on the fission of the transfer products of $^{238}$U projectiles, impinging on a $^{12}$C target, have been performed at GANIL, Caen, in inverse kinematics, covering fission probabilities [117] and fission-fragment properties like the yields of all produced nuclides that are identified in Z and A, as well as fission-fragment kinetic energies [168].

The most remarkable enlargement in the number of systems being accessible for low-energy fission studies was provided by exploiting the fragmentation of relativistic $^{238}$U projectiles at GSI, Darmstadt. From the results obtained up to now, one can estimate that more than 100 mostly neutron-deficient projectile fragments with $A \leq 238$ are accessible for low-energy fission experiments in inverse kinematics by electromagnetic excitations [116, 171, 192, 258]. When fission events after nuclear interaction are suppressed, the induced excitation-energy distribution centers at about 14 MeV above the ground state with a FWHM of about 7 MeV [258]. The first results have provided a systematic mapping of the transition from asymmetric fission around $^{239}$U to symmetric fission around $^{217}$At by measuring the fission-fragment Z distributions [258].

Recently, investigations with relativistic secondary beams were also performed at GSI by the SOFIA collaboration (studies on fission with Aladin) with an improved set-up that provided an unambiguous identification of the fission fragments in Z and A [116, 171, 192]. An exploratory study revealed that this method is also applicable to lighter nuclei, extending to the neutron-deficient lead region [171].

### 3.2. Boosting the fission-fragment kinetic energies

The identification of fission products poses a severe problem. The first experiments were based on radiochemical methods [259, 260], and although this approach provides unambiguous nuclide identification (in A and Z) of the fission fragments, it is not fast enough to determine the yields of short-lived fragments, and it suffers from normalization problems, e.g. by uncertainties of the gamma-spectroscopic properties. Identification with kinematical methods by the double time-of-flight [261, 262] and double-energy measurements [263] provides complete mass distributions, however, with a limited resolution of typically four mass units FWHM [115] and problems in the mass calibrations due to uncertainties in the correction for prompt-neutron emission [126]. At the expense of a very small detection efficiency, the COSI-FAN-TUTTE set-up [264, 265] had some success in measuring the mass and nuclear charge of fission fragments at high total kinetic energies in the light group of thermal-neutron-induced fission of some suitable target nuclei by combining double time-of-flight, double-energy and energy-loss measurements. The LOHENGRIN separator brought about big progress in identifying the fission products in mass and nuclear charge [266], although the Z identification was also limited to the light fission-product group. This technique was applied to the thermal-neutron-induced fission of a number of suitable targets, which were mounted on the ILL high-flux reactor, see [236]. Recent attempts to develop COSI-FAN-TUTTE-like detector assemblies with higher detection efficiency and better resolution are presently being made, but have showed only limited success up to now [267–271]. In particular, the Z resolution is severely impeded by straggling phenomena.

The full nuclide identification (in Z and A) of all fission products has only been achieved by boosting the product energies in inverse-kinematics experiments and by using powerful magnetic spectrometers [116, 138, 171, 192, 258, 272].

### 3.3. Results

Some of the most prominent new results have been obtained in fission experiments performed in inverse kinematics of electromagnetic-induced fission at relativistic energies [171, 192, 258] at GSI, Darmstadt, and on transfer-induced fission at energies slightly above the Coulomb barrier [168] at the VAMOS spectrometer of the GANIL facility.

In [258], the fission-fragment Z distributions of 70 fissionable nuclides from $^{205}$At to $^{234}$U were measured, using the beams of projectile fragments produced from a 1 A GeV $^{238}$U primary beam and identified by the fragment separator of GSI, Darmstadt. The measured Z distributions show a gradual transition from single-humped to double-humped distributions with increasing mass, with triple-humped distributions for fissioning nuclei in the intermediate region around $A = 226$. The position of the heavy component of asymmetric fission can be followed over long isotopic chains and turns out to be very close to $Z = 54$ for all systems investigated. In a refined analysis, it was shown that the mean Z values of the contributions to the heavy component from the two most prominent asymmetric fission channels are nearly the same for all actinides [273]. Moreover, the odd–even structure in the Z yields was found to systematically increase with the asymmetry and to have similar magnitudes for even-Z and odd-Z fissioning nuclei at large asymmetry [274, 275]. The importance of these findings for
the theoretical understanding of the fission process is further discussed in sections 4.3.2 and 4.3.3.

The SOFIA experiment [116, 171, 192], which used a refined and extended set-up compared to the one used in [258], made it possible to fully identify unambiguously and event-by-event all the fission products in Z and A from the electromagnetic-induced fission of the relativistic 238U beam and its fragmentation residues. The experiment also profited from the higher available beam intensity, which allowed the range of nuclei being investigated to be extended and reduced the statistical uncertainties.

As one of the most prominent results, this experiment showed for the first time that the fine structure in the fission-product N distribution depends only weakly on the excitation energy of the fissioning system, in contrast to the odd–even staggering in the Z distribution. This can be seen in figure 1, where the logarithmic four-point differences of

\[ \delta_N(N + 3/2) = 1/8 (-1)^N + 1 \left( \ln Y(N + 3) - \ln Y(N) - 3[\ln Y(N + 2) - \ln Y(N + 1)] \right), \]

of the fission-product N distribution from the SOFIA results for the electromagnetic-induced fission of 238U are compared with those obtained for the thermal-neutron-induced fission of 235U. Around the maximum of the N distribution at \( N \approx 56 \), the \( \delta_N \) values are almost identical, and they are fairly close below and above

\[ N = 56. \]

In contrast, the odd–even staggering in the Z distribution decreases by about 50% [275] when comparing electromagnetic-induced with thermal-neutron-induced fission; see section 4.3.3 for further discussion of the fine structure in the fission-fragment yields.

In the VAMOS experiment on transfer-induced fission and fusion–fission around the Coulomb barrier in inverse kinematics, a separation in Z and A of all fission products has also been obtained [138], although the peaks showed some overlap, preventing an unambiguous event-by-event identification. This experiment provided for the first time complete fission-product nuclide distributions after the formation of a 250Cf compound nucleus at an excitation energy as high as 45 MeV, produced in the fusion of 238U projectiles with 15C [138] and a number of transfer products [281]. This enabled the systematic study of the dependence of the fission-fragment N/Z degree of freedom (charge polarization and fluctuations) on the excitation energy for the first time [138, 165, 281], considering that full nuclide identification had only previously been obtained for the light fission products from the thermal-neutron-induced fission of a small number of fissioning systems.

The observation of a double-humped mass distribution in the fission of the very neutron-deficient 189Hg nucleus in beta-delayed fission and a different kind of structure in the mass distributions in the fission of other nuclei in this region in different experiments [252] demonstrated that complex structural effects are a rather general phenomenon in low-energy fission, not restricted to asymmetric fission in the actinides and multi-modal fission around 258Fm. This result demonstrates that contrary to the symmetric fission in neutron-rich Fm isotopes, which is explained by the simultaneous formation of two fragments close to the doubly magic 132Sn, the production of two semi-magic 90Zr fragments is not favored in the case of 189Hg. A large variety of neutron-deficient nuclei reaching down even below mercury is also accessible to fission studies at energies close to the fission barrier with the SOFIA experiment. A few exploratory measurements have already been made [282].

The experimental observations in the lead region were related to the peculiarities due to shell effects in the potential-energy landscape, calculated with the macroscopic–microscopic approach [23, 29, 154] or with microscopic self-consistent methods [26, 27, 38, 154], partly applied in full calculations of the mass distributions with the scission-point model [23, 26, 29, 66] or in systematic calculations with the stochastic random-walk formalism [56, 154, 252].

A glance at the present status of experimental fission research is exhibited in figure 2, and gives an overview of the observed fission-fragment mass and nuclear-charge distributions. These are among the most prominent signatures of nuclear structure in low-energy fission. Although the coverage on the chart of the nuclides is still far from complete, the systematic variations of the fission-fragment distributions with the mass and nuclear charge of the fissioning system are already rather well in evidence. The experiments performed during the last two decades account for a large share of this achievement.
4. Theoretical innovations

In the following, we will give a survey of the ability of the different presently used theoretical approaches and ideas to closely reproduce experimental fission observables and to reveal the physics behind them. Because the dynamics of the fission process and the influence of shell effects and pairing are considered to be essential assets for understanding low-energy fission, static and purely macroscopic approaches are not included. The survey comprises microscopic self-consistent approaches, stochastic models and a recently developed semi-empirical approach. In contrast to the self-consistent and the stochastic approaches, which are presently restricted to the description of a rather small subset of fission observables, this semi-empirical model describes essentially all fission quantities and their mutual correlations. This is why the physics background and the abilities of this model will be described in more detail in this review.

The self-consistent microscopic theory aims to describe the fission process in a quantum-mechanical framework under the influence of an effective nuclear force, see section 4.1. This is the most ambitious and the most fundamental approach, but it still faces considerable technical and computational difficulties. Also, several formal or conceptual issues have not fully been solved. A review on the present status of this type of model, in particular the implementation, solution methods and results of microscopic self-consistent fission modeling based on the nuclear-density-functional formalism, is given by Schunck and Robledo [81].

Stochastic models describe the fission process by a transport equation, whereby the concepts of statistical mechanics are applied to describe the irreversible approach towards statistical equilibrium, see section 4.2. In recent applications, the stochastic processes are introduced by a random force in the numerical solution of the Langevin equations or by a random-walk approach. These stochastic models are essentially classical, while quantum-mechanical features only enter partially, e.g. by the macroscopic–microscopic description of the potential-energy landscape. Detailed considerations on the validity of transport theories and a survey of the respective theoretical approaches can be found in [285, 286].

A recently developed semi-empirical model description by-passed the great complexity that is encountered when striving for the ab initio modeling of the fission process by concentrating on the essential features of the process that mostly influence the observables, see section 4.3. Qualitative and semi-quantitative ideas in this direction were developed a long time ago, see for example [288]. The ability to obtain rather accurate consistent quantitative results for practically all fission observables was achieved by linking the observables

Figure 2. An updated overview of fissioning systems investigated up to ~2016 in low-energy fission with excitation energies up to 10 MeV above the fission barrier. In addition to the systems where fission-fragment mass distributions (FFMDs) were previously obtained in particle-induced and spontaneous fission (○), the nuclei for which the fission-fragment Z distributions after electromagnetic excitations were measured in an experiment in 1996 [258] and in the recent SOFIA experiment [171, 192] in inverse kinematics at the FRS at GSI (×) and the fissioning daughter nuclei studied in β-delayed fission (⋄) are also shown. Full diamonds mark systems produced after beta decay for which the FFMD were measured; the data are from [154, 252] and the references therein. Furthermore, 25 nuclei are marked (+), including the FFMDs obtained from multi-nucleon-transfer-induced fission with the 18O+232Th [183] and 18O+238U [255] reactions. Several examples of measured FFMDs are shown; data from [254, 278, 283, 284]. For orientation, the primordial isotopes are indicated by squares. Adapted from [249]. © IOP Publishing Ltd. All rights reserved.
for different conditions (for example for fissioning systems with different A and Z and for different initial excitation energies and angular momenta) with a tailored theoretical frame. Thus, the experimental data were traced back to a rather limited number of about 100 empirical model parameters, from which about 50 are decisive for the actual fission process. These simultaneously describe a large variety of fissioning systems with a unique set of parameter values.

4.1. Microscopic self-consistent approaches

4.1.1. Basic considerations. A number of dynamical self-consistent quantum transport theories have been developed for handling nuclear reactions ranging from ab initio to self-consistent mean-field approaches. A description of the different methods can be found, for example, in the habilitation thesis of Lacroix [289]. The application to nuclear fission poses considerable challenges to suitable algorithms and computation resources and is presently an active field of development. Up to now, only theories based on the mean-field approximation have been applied to fission. Due to the tremendous number of possible final configurations, the fissioning system must be treated as an open system, where only a sub-class of the degrees of freedom associated with the system is explicitly considered, because it is technically impossible to simultaneously treat all (collective and intrinsic) degrees of freedom explicitly in a fully quantum-mechanical framework [62, 290]. Either only a few collective degrees of freedom are treated, or the collective degrees of freedom result from the assumption of non-interacting nucleons in a mean field. Moreover, microscopic theories also suffer from an incomplete treatment of nuclear dissipation. Techniques have been proposed to model quantum dissipation in collective motion as an interaction with a heat bath that represents all intrinsic degrees of freedom, e.g. [291–293], but also this simplified scenario has not yet been used to model fission. In general, current microscopic fission models, which will be described in the following, either assume adiabaticity (the decoupling of collective and intrinsic degrees of freedom), or they only include one-body dissipation. Two-body dissipation is only partially treated by including pairing correlations in the calculations that consider one-body dissipation.

4.1.2. Density-functional theory. As said before, all the existing microscopic models of fission are based on the mean-field approximation. Unfortunately, there is significant ambiguity in the names given to the different approaches in the literature. Therefore, for the sake of clarity, here we adopt the notation given in the recent review article on the microscopic theory of fission by Schunck and Robledo [81].

As shown in [81], all the microscopic models that have been applied to fission so far make use of the density-functional theory (DFT), where the term DFT includes self-consistent mean-field theory and extensions beyond the mean field. Within this framework, many-body nucleon interactions are approximated by a mean field, where the nucleons evolve independently, the mean field itself being determined by the ensemble of nucleons. The energy of the nucleus is a functional of the one-body density matrix, which is determined by solving the Hartree–Fock (HF) or the Hartree–Fock–Bogoliubov (HFB) equation, although in some cases the pairing is introduced by using the BCS approximation to the HFB equation. Introducing an energy-density functional (EDF) [294] offers considerable conceptual and practical simplifications to the solution of the many-body problem. In general, the EDF is derived from an effective two-body nuclear potential, typically the Skryme or the Gogny effective interactions. Note that the parameters of these effective interactions are adjusted to the experimental data. For attempts to deduce the nuclear force from QCD see [295, 296].

Fission dynamics can only be treated approximately, either by assuming adiabaticity, i.e. no coupling between collective and intrinsic degrees of freedom, or by representing the wave function at each time by the solution of an HF, or, only very recently, an HFB mean field. We refer to [81] for a detailed description of the DFT theory applied to fission and a complete list of the appropriate references. Here, we only intend to give an overview of the achievements of this kind of approach in contributing to a better understanding of the fission process, on the variety of sophistication and the challenges self-consistent microscopic fission theory is facing.

4.1.3. Application to fission. Table 1 lists the main approaches based on the DFT that have been applied to fission, together with their main assumptions as well as the fission quantities and fissioning systems that have been considered. Notice that the study of odd-mass fissioning nuclei poses additional difficulties with respect to the even–even nuclei, and thus they are only rarely considered in DFT.

The first two approaches listed in table 1 have a first static step in common, where the potential-energy surface (PES) as a function of a few selected collective coordinates and the collective inertia, which determines the response of the fissioning nucleus to changes in the collective variables, are calculated by solving the HFB equation. Fission dynamics is considered as a second step. For computing the PES, the energy of the system is determined under the constraint of the coordinates of the relevant degrees of freedom on a grid of points. At each point, the shape of the system is optimized in a self-consistent way with respect to its energy while respecting the imposed constraints. Figure 3 shows the potential energy surface of $^{232}$Th, calculated using a self-consistent mean-field approach, as an example.

The choice of the collective variables is a key point in these approaches. Reasonable choices of the relevant collective variables (often coming from studies performed with the semi-classical models described in section 4.2) are usually made, but there is always some degree of arbitrariness in the selection. The addition of collective variables or changes in the definition of the relevant collective degrees of freedom can significantly reduce the discontinuities in the PES [297] and strongly impact the results. We further discuss the issues of the number of collective coordinates and the PES discontinuities below and in section 5.1.
A synthetic description of the main microscopic approaches that have been applied to fission.

| Theory | Main assumptions | Quantities and systems described | Selected references |
|--------|------------------|----------------------------------|---------------------|
| Density-functional theory with energy-density functionals based on the Skyrme or Gogny effective potential | Description of the potential-energy surface with a few selected collective degrees of freedom. 
Tunnel transmission coefficient obtained with the WKB approximation. | Spontaneous-fission half-lives of even–even fissioning nuclei | [45, 305, 34] |
| | Description of the potential-energy surface with a few (presently two) selected collective degrees of freedom. The scission configuration must be explicitly defined. Dynamical evolution from the ground state to scission obtained with TDGCM-GOA assuming adiabaticity, i.e. no coupling between collective and intrinsic degrees of freedom. | Fission-fragment mass and charge distributions of few even–even fissioning nuclei, mainly $^{235,237}$Th, $^{236,238}$U, $^{240}$Pu and $^{252}$Cf at low energies. | [299, 306, 77, 96, 98] |
| | Dynamical evolution from a point beyond the fission barrier with TDHF, TDHF-BCS or TDHFB. This implies: 
– Semi-classical evolution of one-body collective degrees of freedom. No tunneling 
– One-body dissipation is included. TDHFB and TDHF-BCS also partly include the Landau–Zener effect. | Most probable kinetic and excitation energies of the fission fragments for a few even–even fissioning nuclei, mainly $^{239}$Pu at low energy and $^{256,260}$Fm, spontaneous fission. Only recently have fairly realistic kinetic-energy and fragment-mass distributions been obtained for $^{256}$Fm. | [47, 51, 59, 62, 68, 71, 95] |

**Note.** All approaches are based on the density-functional theory, as described in [81]. The second column lists the main assumptions underlying the different approaches, the third column gives the fission quantities and fissioning systems that have been computed, and the last column gives some selected references for each approach.

The first approach listed in table 1 is mainly used in spontaneous fission, where the evolution of the nucleus from the ground state to scission proceeds by tunneling through the fission barrier. Spontaneous fission is generally characterized by the fission half-life, which is inversely proportional to the transmission coefficient through the multidimensional PES. The transmission coefficient is computed semi-classically with the Wentzel–Kramers–Brillouin (WKB) approximation [298]. From the calculated PES, it is possible to deduce a one-dimensional fission barrier, which can be compared to the one-dimensional barriers deduced from the experimental data on fission cross sections or probabilities; this will be discussed in section 4.3.1.

The second approach in table 1 is generally applied to induced fission at low energies. The evolution of the fissioning nucleus is computed using the adiabaticity approximation. As said above, this implies that the collective coordinates are completely decoupled from the intrinsic degrees of freedom, and, consequently, the system does not experience any intrinsic excitation during the course of the reaction. The dynamical evolution up to scission is obtained with the time-dependent generator coordinate method TDGCM [77, 243, 299], assuming the Gaussian overlap approximation GOA [300]. A step towards a full dynamical microscopic description of the fission process that goes beyond the adiabatic approximation is a generalization of the TDGCM approach by including two-quasi-particle excitations on the whole fission path in the Schrödinger collective intrinsic model by Bernard et al [15]. To our knowledge, this approach has not yet been applied to infer measurable fission quantities. Within the adiabatic approximation, there is no scission mechanism, and there is again some part of arbitrariness in the definition of the scission configuration. The definitions of scission are either geometrical (e.g. the neck size) or dynamical (the ratio between the Coulomb and the nuclear forces). These definitions ignore the quantum-mechanical effects in the neck region. These effects were investigated by Younes and Gogny [22], who observed that the fission-fragment density distributions have large tails that extend to the other fragment, even when the size of the neck is very small. The authors extended the quantum-localization method used in molecular physics to fission by introducing a localization indicator to sort the quasi-particles into the two pre-fragments. They showed that the application of quantum localization yields more realistic fission-fragment properties at scission. The quantum-localization approach was extended to a finite temperature (i.e. excitation energy above zero) by Schunck et al [61]. The latter studies of scission are static in nature. We will show below how the third type of approach can contribute to a better understanding of the dynamics of scission.

In [81], the third type of approach listed in table 1 is grouped in the so-called time-dependent density-functional theory (TDDFT). In the literature, however, these approaches are denoted in different ways: time-dependent Hartree–Fock TDHF, time-dependent Hartree–Fock–Bogoliubov TDHFB or time-dependent energy-density functional TDEDF. In TDDFT, the real-time evolution of the nuclear system is simulated from an initial condition up to scission by calculating the nuclear wave function at each time with a mean field that results from solving the HF or the HFB equation. TDDFT performs the evolution of single-particle states self-consistently.
under the assumption that the system is described by a state of independent particles or a Slater determinant at all times. From this evolution, one can infer the behavior of any collective one-body degree of freedom (as e.g. multipole deformation, neck formation, fragment kinetic energies, etc). However, the assumption of independent particles leads to a strong underestimation of the fluctuations of the collective degrees of freedom. In this sense, one can consider that in TDDFT the evolution of the collective degrees of freedom is treated semiclassically [301]. As explained in [81], this prevents such types of approach from predicting realistic fission-fragment distributions, and, generally, only average fission-fragment properties can be calculated. In addition, tunneling through the barrier is not included, which implies that TDDFT calculations of low-energy or spontaneous fission have to start beyond the fission barrier. The starting point or the initial condition of the calculations is usually obtained following the assumption of independent particles or a Slater determinant at all times.

Moreover, with the exploration of the initial phase space (even without the inclusion of pairing effects) no dynamical threshold was observed. In total, a few hundred trajectories starting from different initial densities were propagated in time. This sampling of trajectories was possible because TDHF BCS is significantly more affordable numerically than TDHFB [81]. Solving the TDHFB equation is substantially more involved numerically than solving the TDHF equation. In [68], the evolution from a point slightly beyond the barrier up to scission was achieved by allowing transitions between magnetic substates by means of a complex pairing field that varies in time and space during the evolution. The evolution of the proton and neutron densities and of the corresponding pairing fields for the fission of $^{240}$Pu at an excitation energy of 8.05 MeV are shown in figure 4.

More recently, Tanimura et al [95] proposed a novel method to describe quantum fluctuations and spontaneous symmetry breaking together with the possibility of obtaining fully microscopic fragment mass and TKE distributions in spontaneous fission. The method consists of simulating quantum and thermal effects by sampling the initial conditions, followed by quasiclassical evolutions with a TDHF + BCS approach. A significant increase of the fluctuations in the collective degrees of freedom by the sampling of initial conditions was observed. Moreover, with the exploration of the initial phase space (even without the inclusion of pairing effects) no dynamical threshold was observed. In total, a few hundred trajectories starting from different initial densities were propagated in time. This sampling of trajectories was possible because TDHF + BCS is much more affordable numerically than TDHFB [81].

4.1.4. Selected results. In the following, we will discuss some selected results obtained with the three approaches listed in table 1. Staszcak et al [305] studied the multi-modal spontaneous fission of isotopes from californium to hassium with DFT solving the HF-BCS equation in two-dimensional collective spaces with the Skyrme effective interaction. They calculated a two-dimensional PES involving the quadrupole and octupole moments and another two-dimensional PES involving the quadrupole and hexadecupole degrees of freedom. Observed fission characteristics in this region were traced back to topologies in the multidimensional collective space by searching for the optimum collective trajectory. The authors predicted tri-modal fission for several rutherfordium, seaborgium and hassium isotopes where the compact symmetric (consisting of two spherical two-body dissipation is partially considered. In the TDDFT, scission occurs naturally at some time as a result of the competition between nuclear and Coulomb forces. Because the fragments can experience a rapid change in shape at scission, the TDDFT is better suited to describing scission dynamics than the adiabatic approach previously described.

In most calculations based on the TDDFT, the starting point is required to be located well beyond the outer barrier (e.g. [51, 59, 62]) or to have an additional initial collective energy or boost (e.g. [62, 71]) for the system to evolve to scission. The appearance of such a dynamical threshold was recently attributed by Bulgac et al [68] to restrictions for transitions of a Cooper-pair state with opposite angular momenta into other Cooper-pair states with different angular momenta. This prevents the system from adopting the most strongly bound states. Bulgac et al were the first to perform a full TDHF calculation of induced fission, which they refer to as time-dependent superfluid local-density approximation TDSLDA. Solving the TDSLDA equation is substantially more involved numerically than solving the TDHF equation. In [68], the evolution from a point slightly beyond the barrier up to scission was achieved by allowing transitions between magnetic substates by means of a complex pairing field that varies in time and space during the evolution. The evolution of the proton and neutron densities and of the corresponding pairing fields for the fission of $^{240}$Pu at an excitation energy of 8.05 MeV are shown in figure 4.

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fragments of the same mass), the elongated symmetric and the elongated asymmetric modes co-exist. The spontaneous-fission half-lives were calculated with the WKB approximation for the double-humped potential barrier, assuming a one-dimensional tunneling path along the elongation degree of freedom.

In a later work, Staszczak et al [34] solved the HFB equation with the Skyrme effective interaction and computed the spontaneous-fission half-lives along the entire Fm isotopic chain reproducing within 1–2 orders of magnitude the experimental trend of the lifetime as a function of the neutron number of even–even isotopes. Geometrical degrees of freedom, typically multipole moments, are very important for a realistic description of fission. However, recent work showed that fission can also be very sensitive to non-geometric collective variables such as pairing correlations. For example, Sadhukhan et al [45] demonstrated that the inclusion of pairing collective degrees of freedom has a huge impact on the spontaneous-fission half-lives of $^{264}$Fm and $^{240}$Pu.

The first realistic calculations of fission-fragment kinetic-energy and mass distributions obtained with the second approach were performed by Goutte et al [299]. The authors solved the TDGCM equations using the GOA and the D1S Gogny force for $^{238}$U at excitation energies slightly above the fission barrier. The quadrupole and octupole moments were considered to be the two relevant collective parameters of the fissioning system. Figure 5 shows the evaluation [307] for the fission-fragment mass distribution of $^{238}$U in comparison with the results of a static one-dimensional calculation of collective stationary vibrations along the sole mass-asymmetry degree of freedom (left-right asymmetry) for nuclear configurations just before scission and with a full time-dependent calculation involving the two-dimensional PES. The maxima of the one-dimensional mass distribution lie close to the Wahl evaluation, indicating that the most probable fragmentation is essentially due to the properties of the potential-energy surface at scission, i.e. to shell effects in the nascent fragments. However, it is clear that reproducing the width of the mass distribution requires the inclusion of dynamical effects in the descent from saddle to scission of the two-dimensional PES.

The same approach as Goutte et al was used by Younes and Gogny [306] to compute the fission-fragment mass distributions of $^{236}$U and $^{240}$Pu at different neutron incident energies ranging from 0 to 5 MeV. Instead of the commonly used multipole moments, Younes and Gogny used two collective variables similar to the ones used in the macroscopic–microscopic approaches: the distance between the fragments and the mass asymmetry. As a result of this choice, the discontinuities in the PES were significantly reduced, particularly near scission. Figure 30 in [81] shows that the resulting mass distributions of $^{240}$Pu agree fairly well with the data.

Regnier et al [77] performed an elaborate study of the influence of using different effective forces and different initial conditions on the pre-neutron mass and Z distribution in the neutron-induced fission of $^{239}$Pu at $E^* = 1$ MeV above the fission barrier with the adiabatic TDGCM-GOA method. They found that the qualitative features of the fission-fragment mass distributions are rather robust with respect to the effective force (see figure 6) and/or the various ingredients of the model. The calculated mass distributions agree rather well with the experimental data, although there remain ambiguities in deducing the widths of the dominating asymmetric components from the calculation due to difficulties in the modeling of the fluctuations.

Calculations for the fission of $^{252}$Cf were performed by Zdeb et al [98] using the TDGCM-GOA approximation with the aim of examining how the initial conditions affect the obtained fission-fragment mass yield—especially how this quantity depends on the excitation energy and the parity of the initial state. The dependence of the mass distribution on the initial conditions was found to be rather weak up to initial excitation energies 4 MeV above the fission barrier. Possible modifications of the model are discussed in order to enhance the yields at large asymmetry, which are considerably underestimated.

Recently, the induced fission of $^{229}$Th was investigated with the TDGCM-GOA using a relativistic energy-density functional by Tao et al [96]. The overall topography of the calculated PES and the TKE values was found to be consistent with previous studies based on the Gogny density functional [308]. The calculated charge distribution reproduces the main characteristics of the measured distribution, which presents a triple-humped structure with symmetric and asymmetric peaks. The influence of the strength of the pairing interaction and of the initial excitation energy were studied. Some prominent results are shown in figures 7 and 8. Note that by its nature the model cannot describe the odd–even staggering. The dependence on the excitation energy appears to be rather weak, compared to the experimental systematics [258, 309, 310].
As said above, the approaches based on the adiabatic approximation require a criterion to define the scission configuration. Moreover, close to scission an acceleration of the fragments may occur, inducing non-adiabatic effects. Thus, the adiabatic approximation is expected to break down in the latter stage of fission. These effects are crucial for properly describing the properties of the fragments such as their kinetic and excitation energy. The TDDFT is more appropriate for calculations of these properties since there is no need to characterize scission, and one-body dissipation is included.

Simenel and Umar [47] studied dynamical effects in the vicinity of scission on the kinetic and excitation energies of the fragments in the symmetric fission of $^{264}$Fm with time-dependent Hartree–Fock calculations. As a starting point, they considered the position after the barrier where the properties of the emerging double-magic $^{132}$Sn fragments are well defined. Simenel and Umar showed that a significant fraction of the final excitation energy of the fragments is acquired right before scission, and that it is at least partly stored in the low-energy collective vibrational states of the fragments. However, they neglected pairing correlations, which may be reasonable when studying $^{264}$Fm but not when investigating fission dynamics across the nuclear chart. Their approach was extended by including pairing correlations within the BCS approximation by Scamps et al [59] and was applied to the investigation of the multi-modal fission of $^{258}$Fm. The resulting TKE of the fragments was compared with the experimental data showing good agreement. Their result for the TKE of the compact symmetric mode was located near the main peak of the TKE distribution in agreement with previous interpretations of the experimental data. The measured lower TKE tail was mostly attributed to the elongated asymmetric mode, although these calculations predict that the elongated symmetric mode leads to the lowest values of the TKE where only a few events have been observed experimentally. They showed that the time associated with the non-adiabatic descent of the potential to scission for the compact symmetric mode is shorter than for the two other modes, reflecting that magic fragments are difficult to excite and deform, and, thus, fission occurs faster as less dissipation is involved.

Tanimura et al [62] proposed a method to obtain the collective momentum and mass associated with any collective observable from TDDFT. They applied this method to the fission of $^{258}$Fm and showed how the use of different amounts of collective energy to induce a boost for overcoming the dynamical threshold can impact the results. They also showed that in the non-adiabatic case the fragments stick together for a longer time than in the adiabatic case, indicating that the
scission point derived in the non-adiabatic case occurs at a larger distance between the fragments.

In the first ever application of TDHFB to fission, Bulgar et al. [68] presented rather accurate calculations with the time-dependent superfluid local-density approximation (TDSLDA) of some specific average fission quantities (fragment excitation energies, kinetic energies, saddle-to-scission time) in the low-energy-neutron-induced fission of $^{239}$Pu. The threshold anomaly (the necessity of imposing an initial boost in order to bring the system to fission that was observed in previous studies) is solved by treating the dynamical pairing. Long fission times were obtained that were attributed to the excitation of a large number of collective degrees of freedom (confirming the early qualitative results of Nörenberg [246]) and not to a particularly large viscosity, i.e. coupling between collective and intrinsic degrees of freedom.

Tanimura et al. [95] studied the fission-fragment mass and TKE distributions of $^{258}$Fm spontaneous fission with TDHF-BCS. They simulate quantum and thermal effects by sampling the initial conditions, followed by the quasi-classical evolution of the collective motion with TDDFT. As said above, the authors observed that this exploration of the initial phase space (even without the inclusion of dynamical pairing effects) also solves the dynamical-threshold anomaly. The calculated TKE distribution is in rather good agreement with the experimental data, whereas the mass distribution is still too narrow, see figure 9. From their model Tanimura et al infer information on the fission process. For instance, they show that at scission the fragments are deformed and that the deformation relaxes as the two fragments move apart from each other after scission. In their calculation, the fragments are excited before reaching scission, which leads to a non-negligible probability for pre-scission neutron emission.

TDDFT appears to be the most ambitious self-consistent microscopic approach for the description of the fission process. Therefore, it is expected to gain importance and play a leading role in fission research in the future. As described above, two implementations of this approach have been developed, either using the BCS approximation (e.g. by Tanimura et al. [95]) or the Hartree–Fock–Bogoliubov approximation (by Bulgar et al. [68]). Apparently, some important results are not compatible. Table 2 summarizes some prominent differences in the ingredients, in the application and in the results of the two approaches. The scientific discussion which aims to solve this problem is in progress. A crucial point is the approximate treatment of the pairing correlations within the TDBCS approximation in [95], which, according to [68], violates the continuity equation.

As we have seen, at present, the importance of microscopic self-consistent models for the description of nuclear fission lies in the qualitative understanding of several fundamental aspects, while the achievements in completeness and accuracy are in a vivid process of development. Only a limited variety of fission observables (mostly spontaneous-fission half-lives, fission-fragment mass or nuclear-charge distributions and fragment kinetic energies) for a few fissioning nuclei have been...
4.2. Stochastic approaches

4.2.1. Basic considerations. Early dynamical studies of the nuclear-fission process have been performed by Nix [312] with a non-viscous irrotational liquid-drop model and by Davies et al [313] with the inclusion of two-body dissipation. Later, Adeev et al [314] studied the influence of one-body and two-body dissipation on fission dynamics with a transport equation of the Fokker–Planck type. Also in this case, like in the calculation of the potential-energy surface by microscopic self-consistent approaches, only a limited part of the large number of degrees of freedom was explicitly treated. In [314], the evolution of the probability-density distribution in a space defined by the restricted number of degrees of freedom considered to be relevant is described under the influence of driving force and friction, including the associated statistical fluctuations. Driving force and friction represent the interactions with the other degrees of freedom, which are treated as a heat bath and, thus, are not explicitly considered. However, the solution of the Fokker–Planck equation was limited to simple cases or subject to strong approximations. Abe et al [315] replaced the Fokker–Planck equation by the equivalent Langevin equations, which can be solved numerically in application to fission. Monte-Carlo sampling of the individual trajectories in the space of the relevant degrees of freedom proved to be a more practicable way of obtaining more accurate solutions in complex cases; however, this method does require considerable computing resources.

4.2.2. Stochastic transport equations. The Langevin equations in their discretized form for the evolution of the system in the time interval $\Delta t$ between the time step $i$ and $i + 1$, read

\[
q_{i+1} = q_i + \frac{p_i}{m} \Delta t + \sqrt{\beta m T \Delta t} \cdot \Gamma.
\]  

(2)

$q$ is the the vector of coordinates that denotes the position in the space of the relevant degrees of freedom, $p$ is the corresponding momentum, $m$ and $\beta$ are the mass parameter and the dissipation coefficient, respectively. $-T \frac{\partial \rho}{\partial q}$ is the driving force, $-\beta p_i$ is the friction force, and $\sqrt{\beta m T \Delta t} \cdot \Gamma$ is the fluctuating term that expresses the stochastic transfer of thermal energy between the heat bath and the collective coordinates. $T$ is the temperature and $S$ is the entropy. Both are related to the local level density $\rho$, which has to be provided by an appropriate model, see for example [316].

\[
T = \left( \frac{\text{dln}(\rho)}{\text{d}E} \right)^{-1}
\]  

(3)

and

\[
S = \text{ln}(\rho).
\]  

(4)

In most practical cases, the stochastic variable $\Gamma$ that defines the fluctuating force is linked to the dissipation strength by the fluctuation-dissipation theorem [317].

Equations (1) and (2) are valid, if $m$ and $\beta$ do not depend on the coordinates $q$ and the direction of motion. In the general case, $m$ and $\beta$ are tensors, and the Langevin equations must be adapted. At low energies, pairing correlations and shell effects should be included not only in the potential-energy surface, but also in the level density [97], in the friction tensor [39] and in the mass tensor [318]. However, this is rarely done. For more detailed information on the application of the Langevin equations to fission and to other nuclear reactions we refer to the review article of Frübrich and Gontchar in [319].

Since a nucleus is an isolated system with a fixed total energy and fixed particle number, equations (1) and (2) must be formulated in the fully micro-canonical version (specified as option 1 in table 3). This entails that the temperature $T$ and entropy $S$ are given by the level density with equations (3) and (4), respectively, at an energy equal to the total energy of the system minus the local potential and the actual collective energy. This way, the potential energy does not explicitly appear in equation (2), but its influence is felt via temperature $T$ and entropy $S$.

Strictly speaking, the degeneracy of the magnetic sub-states should be considered.

| Approach                        | TDHF-BCS, [95]                        | TDSLDA, [68]                        |
|--------------------------------|--------------------------------------|-------------------------------------|
| Pairing model                  | BCS approximation                     | Bogoliubov theory                   |
| Starting point of the dynamic calculation | Between saddle and scission with thermal fluctuations | Near outer saddle                  |
| Main influence on fission time  | Dissipation (intrinsic excitations)   | Excitations of collective shape and pairing modes |
| Typical fission time            | 1500 fm c$^{-1}$                      | 10000 fm c$^{-1}$                   |
| Disappearance of threshold anomaly mainly attributed to | Fluctuations of the fission path | Dynamical pairing                   |
This is important in applications to low-energy fission, where approximations that are often applied (specified as options 2–4 in table 3) only badly represent the statistical properties of the nucleus. Also, the influence of pairing correlations and shell effects on the binding energy and the level density should be properly considered, in particular at low excitation energies. This is not so critical at higher excitation energies, where for example the use of Boltzmann statistics may be a suitable approximation. These aspects have been stressed in several places, for example by Fröbrich in [320]. He also stresses that the driving force is not given by the derivative of the potential, but by the derivative of the entropy times the temperature that expresses the influence of the environment on the selected degrees of freedom according to the laws of statistical mechanics, see equation (2). Due to the complexity of the nuclear level density, this can lead to very different results.

If very strong friction is assumed, the motion becomes over-damped, and the influence of the mass tends to vanish. This case is represented by the Smoluchowski equation [319], which requires less computational expense. As argued in [18], even less demanding in computing resources is the replacement of the kinematic equations (1) and (2) with a random-walk approach using Metropolis sampling [321]. All these different approaches are presently in use.

4.2.3. Application to fission. In practice, the application of stochastic classical approaches is performed in two steps, like in the case of the first two self-consistent microscopic approaches listed in table 1, where a small number of collective degrees of freedom are explicitly considered. In the first step, the potential energy is computed on a grid in the space determined by the relevant degrees of freedom, usually by the macroscopic–microscopic model. In most cases, the relevant degrees of freedom are the coordinates of a suitable shape parametrization. Eventually, the potential energy is minimized individually on each grid point with respect to some additional shape parameters. Also, the dissipation tensor and the mass tensor must be defined, for example on the basis of one-body and two-body dissipation with phenomenological adjustments and the hydrodynamical inertia with the Werner–Wheeler approximation for the velocity field [313], respectively. With these ingredients, Monte-Carlo sampling of the fission trajectories with one of the stochastic approaches listed in table 3 is performed.

4.2.4. Selected results. To our knowledge, stochastic approaches have been applied to low-energy fission with the inclusion of shell effects since 2002. Ichikawa et al [322] studied the fission of 270Sg with three-dimensional Langevin calculations at an excitation energy of 10 MeV above the ground state. By considering the energy dependence of the shell effect, the calculation was essentially microcanonical. The shell effects were obtained with the two-center shell model [323]. The mass tensor was calculated using the hydrodynamical model with the Werner–Wheeler approximation [324] for the velocity field, and the wall-and-window one-body dissipation [325] was adopted for the dissipation tensor.

The distance of the fragment centers, the quadrupole deformation—assumed to be common to both fragments—and the mass asymmetry were chosen as shape parameters. The measured mass distribution was well reproduced, while the total kinetic energy (TKE) was overestimated. The authors stressed the strong influence of the dynamics on the mass distribution. This model has been applied in [326] to study the multi-modal fission of 256,258,264Fm. In [327], the influence of the dissipation tensor on the fission trajectory was demonstrated.

Aritomo et al [328] succeeded in reproducing fairly well the measured fission-fragment mass distributions and the TKE distributions of 235U, 236U and 240Pu before prompt-neutron emission at an excitation energy of 20 MeV with their stochastic approach, similar to the one applied before in [322, 326, 327] and using the same shape parametrization. They failed to reproduce the transition to a single-humped mass distribution towards 226Th and 222Th, and attributed this to an insufficiently detailed shape parametrization. In particular, they concluded that the deformation parameters of the two nascent fragments should be chosen independently. In [35], Aritomo et al introduced a new shape parametrization, but stuck to three dimensions. They tested the model against the mass-TKE distribution for the fission of 256U at an excitation energy of 20 MeV. The influence of pairing correlations that may be assumed to be weak at this energy is neglected. In [49], Aritomo et al considered the N/Z degree of freedom in their model, which enabled them to calculate independent yields, which means the fission-fragment yields specified in A and Z.

A comprehensive database of five-dimensional potential-energy landscapes, calculated by Möller et al [329] with the macroscopic–microscopic approach, was used by Randrup et al as a basis for the wide-spread stochastic calculations of pre-neutron fission-fragment mass distributions [17, 18, 25, 32]. The Z distributions were deduced with an unchanged charge-density (UCD) assumption, which means that the N/Z of the fragments is equal to that of the fissioning nucleus. Due to the relatively large number of five shape parameters (overall elongation, constriction, reflection asymmetry and deformations of the two individual pre-fragments), some simplifications and approximations in the dynamical treatment were applied in order to keep the computational needs to an

| Name                                      | Approximations                                      |
|-------------------------------------------|-----------------------------------------------------|
| Langevin equations, micro-canonical        | Classical dynamicsa                                 |
| Langevin equations, not fully micro-canonicalb | Classical dynamicsa + simplified driving force or state density |
| Smoluchowski equation                     | Classical dynamicsa + over-damped motion            |
| Random walk                               | Classical dynamicsa + over-damped motion + Metropolis sampling |

a Certain quantum-mechanical features can effectively be considered in the classical Langevin equations, for example shell effects in the potential energy, the contribution of the zero-point motion to fluctuation phenomena, etc.
b Different kinds of approximations, for example, the coupling to a heat bath of constant temperature, Boltzmann statistics, etc.
affordable level. A random-walk approach using Metropolis sampling was applied, assuming over-damped motion, and the driving force was taken as the derivative of the potential. These approximations prevent realistic results on the energetics of the fission process from being obtained (for example kinetic and excitation energies of the fragments, prompt-neutron multiplicities, etc). The measured mass distributions of a large number of systems, reaching from $^{180}$Hg to $^{240}$Pu, are fairly well reproduced. These results have already been depicted and were duly acknowledged in [249]. The importance of a sufficiently detailed shape parametrization for the calculation of fission-fragment mass distributions, in particular the freedom for the nascent fragments to take individual deformations, is demonstrated. Recently, a method of extending this approach to six dimensions by including the $N/Z$ (charge polarization) degree of freedom has been proposed [55]. In the most recent version of this approach [97], the Metropolis walk is not driven by the potential energy any more, but by the number of available levels. This means that the driving force is calculated microcanonically. By using microscopic level densities, it is possible to study the evolution of the fission-fragment mass distribution as a function of initial excitation energy.

In a recent study [89] on the basis of the Brownian shape-motion model with its recent extensions, the evolution of fission-fragment charge distributions with the neutron number of the compound-system sequence $^{234}$U, $^{236}$U, $^{238}$U and $^{240}$U was studied and compared with the experimental data. The evolution of the location of the peak charge yield from $Z = 54$ in $^{234}$U towards $Z = 52$ in heavier isotopes, seen in the experimental data, was reproduced fairly well. Moreover, it was shown that it is necessary to take multi-chance fission into account to describe the yields already at an excitation energy of 20 MeV. The contributions of the different chances to the fission-fragment $Z$ distribution of the system $^{235}$U(n,f) at $E_x = 14$ MeV, and a comparison of the total distribution with the empirical data is shown in figure 10. The absence of an odd–even effect in the empirical data can be understood by the limited experimental $Z$ resolution.

Ishizuka et al [87] developed a four-dimensional Langevin model, which can treat the deformation of each fragment independently, and applied it to the low-energy fission of $^{238}$U. The transport coefficients were calculated by macroscopic prescriptions. Note that the deformation parameters of the two complementary fragments are set equal in most versions of the stochastic fission approach, with only few exceptions, e.g. in the Brownian shape-motion model [17]. The choice of collective variables, in particular the independent variation of the deformation of the two complementary fragments, allowed them to perform a multi-parametric correlation analysis among the three key fission observables: mass, TKE and prompt-neutron multiplicity.

A strong correlation is found between the mass-dependent deformation of fragments at the scission point and the sawtooth structure of prompt neutron multiplicity, including their dependence on the excitation energy. In detail, the mass-dependent shapes of the complementary nascent fragments at scission develop in opposite directions as a function of the mass asymmetry. The underlying feature—the saw-tooth shape of the mass-dependent prompt-neutron multiplicities—has been known from experiment for a long time (e.g. [332]), and has been investigated by different theoretical approaches, for example by the scission-point model of Wilkins et al [248] or by the random-neck-rupture model of Brosa et al [244]. This demonstrates the importance of considering the shape parameters of the two fragments independently.

Figures 11 and 12 show the calculated fission-fragment mass distributions from [87] for $^{236}$U ($E^* = 20$ MeV) and $^{258}$Fm ($E^* = 7.5$ MeV) in comparison with the experimental data. The influence of the type of potential well—parabolic (TCSM) or Woods–Saxon (TCWS)—and of the dimensionality—3D or 4D—of the Langevin calculation is demonstrated. In particular, the mass-dependent TKE (figure 12) is much better reproduced by the four-dimensional calculation.

The mass distributions and the total kinetic energy of fission fragments for a series of actinides and Fm isotopes at various excitation energies were studied within the three-dimensional Langevin approach by Usang et al [94]. The experimental results are fairly well reproduced. In particular, the obtained variation of the TKE in the neutron-induced fission of $^{235}$Pa, $^{235}$U and $^{239}$Pu follows the experimental trend up to incident-neutron energies of 45 MeV, when microscopic transport coefficients are used, although the calculation was restricted to first-chance fission. This agreement seems to be in conflict with the results from other models (for example, the Brownian shape-motion model [89] or the semi-empirical GEF model [228]), where a strong influence of multi-chance fission was observed for fission-fragment distributions [89, 228] and kinetic energies [228] at excitation energies above the threshold for second-chance fission.

Very recently, Sierk performed calculations with the Langevin approach on a five-dimensional potential-energy surface [93]. The approach is not fully microcanonical, since the driving force is determined by the derivative of the potential. Because dissipation is explicitly considered by the surface-plus-window dissipation model, not only fission-fragment mass distributions, but also total kinetic energies are obtained for the neutron-induced fission of $^{233,235}$U and $^{239}$Pu from thermal energy to a few MeV, as well as for the spontaneous fission of $^{240}$Pu and $^{252}$Cf. The measured pre-neutron mass distributions are considerably well reproduced, while the kinetic energies of the fragments close to symmetry are overestimated. The post-neutron fission-fragment mass distributions acquire the measured widths by assuming a random neck rupture and after a few corrections of the model. Sierk [93] offers a detailed discussion of the model assumptions, approximations and uncertainties, and of possible reasons for the observed deviations from the experimental data.

At present, systematic calculations of fission-fragment mass distributions for a large number of different fissioning systems have only been performed with the simplified dynamics of the Metropolis sampling [56]. The systematic calculations of fission-fragment mass distributions, total kinetic energies, spectra and mass-dependent multiplicities of prompt neutrons, and other fission observables for many systems as a function of initial excitation energy with a fully micro-canonical Langevin approach appear to be possible, but they have not yet been reported. Although this stochastic approach misses

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Note: For clarity, the text has been reformatted to improve readability. The original page numbers and references (e.g., [89, 228], [93]) have been kept intact for context.
the full inclusion of quantum-mechanical features, such results would be very interesting, because these calculations are still out of reach for microscopic, self-consistent models.

4.3. General fission model GEF

4.3.1. Basic considerations. Very recently, an approach to fission that exploits some rather general theorems, concepts and laws\(^8\), combined with empirical information, has been successfully applied to develop a model named GEF (general description of fission observables) [339]. The theorems, concepts and laws serve to replace complex calculations with descriptions that simplify them and, under certain conditions, may even yield more accurate results. Prominent examples are (i) the topographic theorem of Myers and Swiatecki [340], which permits the microscopic calculation of the saddle-point mass to be replaced with a prescription based on a macroscopic model and the empirical ground-state mass, (ii) the application of statistical mechanics to calculate the division of intrinsic excitation energy at scission [12], (iii) the early freeze-out of collective variables discussed e.g. by Asghar [341], and (iv) the early localization of the nucleonic wave functions in the nascent fragments and the manifestation of fragment shells, deduced by Mosel and Schmitt [242] from two-center-shell-model calculations. Such kinds of prescriptions are rather commonly used in nuclear physics. A well-known example is the shell-correction method [298], which replaces the microscopic calculation of the nuclear binding energy with the sum of the smooth part of the total energy, calculated with the phenomenological liquid-drop model, and the shell-correction energy, calculated with the Strutinsky method [342]. The GEF model covers the majority of fission quantities and reproduces the measured observables, especially in the range \(A \geq 230\) — where most of the experimental data were obtained — with a remarkable accuracy that makes it suitable for technical applications. To illustrate this with a few examples, we mention that fission yields from GEF are being used to fill in unmeasured fission-yield data in the JEFF-3.3 evaluation [343]. Moreover, the GEF model was able to reveal that the ENDF/B-VII evaluation for the mass yields of the system \({}^{237}\text{Np}(n_{th},f)\) was based on erroneous data, because the target was contaminated with the contribution of another fissile nucleus, probably \({}^{239}\text{Pu}\) [228]. The accuracy of the GEF calculations can be inferred from the systematic comparison of different fission data with the GEF calculations shown in [228] and some recent publications, where they are

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\(^8\) The term ‘general’ is used here to denote a concept or law of general validity in contrast to \(ab\ initio\) modeling, see for example [337]. Models that are based on general concepts often need empirical information for performing quantitative calculations, while \(ab\ initio\) modeling strives for quantitative results without relying on empirical or fitted parameters. As a critical remark about the sharp separation between these models, we would like to cite Niels Bohr, who stated that ‘it appeared difficult to define what one should understand by ‘first principles’ in a world of knowledge where the starting point is empirical evidence’ [338].
compared to the measured independent [190, 192] and cumulative [344, 345] fission yields and prompt-gamma spectra [212]. The GEF model was also applied to study fragment mass and excitation-energy-dependent prompt-neutron yields [346], and to establish the uncertainty propagation of fission product yields and decay heat [347]. It was also used to estimate the corrected fission-fragment yields, strongly diverging from the ENDF evaluation, in the interest of calculating realistic anti-neutrino spectra for $^{233}$U(n,f) [348]. Large deviations for fission-fragment yields between the experimental data and GEF calculations have been reported in [195, 349]. In the first case, the authors admitted an error in the input options of their GEF calculation [350]; in the second case, the discrepancies may need clarification, if possible by additional measurements with an independent experimental approach.

In the domain of fundamental physics, GEF has been used to interpret measured mass distributions after heavy-ion induced reactions $^{12}$C+$^{235}$U, $^{34}$S+$^{208}$Pb, $^{36}$S+$^{206}$Pb and $^{36}$S+$^{208}$Pb, in terms of multi-chance fusion–fission and quasi-fission [351].

GEF has also proven to be a useful tool for the detailed planning of experimental methods [116, 352]. The calculations of many of the fission quantities that can be obtained from GEF still pose severe problems to stochastic and microscopic models. A detailed documentation of the GEF model code, its underlying ideas and a presentation of a large variety of results can be found in [228, 353, 354]. GEF makes use of several long-standing mostly qualitative ideas that were already able to explain many systematic trends and regularities in several fission observables and combines them with some innovative conceptions. It owes its accuracy and considerable predictive power to the development of additional powerful ideas and the consideration of important experimental findings that were not fully understood before or obtained only recently. In the following, we will describe the most important ideas and their successful application to the GEF model.

4.3.2. Topographic theorem: accurate fission barriers. The modeling of the nuclear-fission process in stochastic and self-consistent models starts with the calculation of the potential energy of the fissioning system in the space defined by the ‘relevant’ collective degrees of freedom. Besides the ground state of the nucleus, the saddle point that defines the fission threshold (the lowest energy at which the system can proceed to fission without tunneling) is a prominent point in the potential-energy landscape. However, in contrast to the nuclear binding energy in the ground state, the binding energy at the fission saddle is not directly measurable. Empirical information on the fission threshold has been derived from measured energy-dependent fission cross sections and/or fission probabilities assuming penetrability through a multi-humped barrier, approximated by one-dimensional inverted parabolas. One of the most comprehensive studies of this type was made in [99]. The resulting values of the barrier parameters depend on the details of the model analysis, for example on the level-density description and, in particular, on the properties of the first excited states above the barriers. Therefore, the empirical fission-barrier heights are considered to be subject to an appreciable uncertainty, usually presumed to be on the order of 1 MeV, see for example [43]. Although the barrier height deduced from this method cannot be identified with the saddle-point energy of multi-dimensional potential-energy surfaces from microscopic or macroscopic–microscopic theories, a link can be established by matching the energy-dependent fission probabilities in the different scenarios. Fission barriers derived using the one-dimensional penetrability approach have a considerable practical importance, because they are very much used to estimate the energy-dependent fission probabilities and neutron-induced-fission cross sections. To our knowledge, the penetrability through a multi-dimensional barrier has not yet been calculated.

In this section, we will derive a well-founded estimation of the uncertainty of fission barriers in the one-dimensional penetrability approach and propose a procedure for predicting accurate fission-barrier values. This is an important information, because it allows a better assessment of the quality of a theoretical model by its ability to reproduce the empirical values of the fission barrier. As we will see, one may draw conclusions on the ability of the model for realistic estimations of the full potential-energy surface of the fissioning system from this result.
Myers and Swiatecki introduced the idea that the nuclear binding energy (or mass) at the fission threshold is essentially a macroscopic quantity [340]. This means that the mass at the highest of the consecutive barriers between the ground-state shape and the scission configuration can be estimated to a good approximation by the saddle mass of a macroscopic model. They deduced this ‘topographic theorem’ from considerations on the topographic properties of a surface in multidimensional space with the specific properties of the potential-energy surface around the fission threshold. The basic idea is illustrated in figure 13, where pairing effects are neglected. The height of the fission barrier \( B_f \) is given to a good approximation by the difference of the macroscopic barrier \( B_f^{\text{mac}} \) and the shell-correction energy in the ground state \( \delta U_{gs} \). In practice, the ground-state shell correction \( \delta U_{gs} \) is determined as the difference of the ground-state energy \( E_{gs-\text{nopair}} \) that is averaged over odd–even staggering in \( N \) and \( Z \) and the macroscopic binding energy: \( \delta U_{gs} = E_{gs-\text{nopair}} - E_{gs}^{\text{mac}} \). If the ground-state mass is experimentally known, the fission barrier can be estimated on the basis of a macroscopic model that provides the macroscopic ground-state mass and the fission barrier:

\[
B_f \approx B_f^{\text{mac}} - E_{gs-\text{nopair}}^{\exp} + E_{gs}^{\text{mac}}. \tag{5}
\]

The condition for this topographic property of a surface in multi-dimensional space is that the wavelength of the fluctuations induced by the shell effects is smaller than the wavelength of the variations induced by the macroscopic potential. This behavior can be understood, because a local modification of the potential by a bump or a dip, for example by the shell effects, does not have a big effect on the height of the saddle. The fissioning nucleus will go around the bump, and it cannot profit from the depth of the dip, because the potential at its border has only changed a little. This phenomenon is related to the observation that the potential at the fission saddle in calculations with a shape parametrization tends to take lower values by allowing for more complex shapes. The inclusion of additional degrees of freedom gives access to a path that is energetically more favorable and avoids the bump mentioned above.

A detailed investigation of the applicability of the topographic theorem was performed in [356]. The validity of the topographic theorem has been demonstrated before in a more qualitative way, for example in [357], and possible explanations for the observed deviations in the range of a few MeV are discussed. The topographic theorem has also been used before as a test of the ability of different theoretical models to describe the long-range behavior of the fission threshold along isotopic chains [358, 359].

According to a previous analysis in [359], the average trend of the saddle mass along isotopic chains is very well reproduced by the Thomas–Fermi model of Myers and Swiatecki [340, 360]. (As will be shown below, this is not the case in the \( Z \) dependence of the saddle mass.) Therefore, the comprehensive set of empirical fission thresholds, which means the maxima of the first and second barrier heights, from [99] that are extracted from the experimental fission probabilities and the cross sections are compared in figure 14 with the quantity

\[
B_f^{\text{topo}} = B_f^{\text{TF}} - E_{gs-\text{nopair}}^{\exp} + E_{gs}^{\text{TF}} \tag{6}
\]

where \( B_f^{\text{TF}} \) denotes the macroscopic fission barrier of [360], represented by \( B_f^{\text{mac}} \) in figure 13, and \( E_{gs}^{\text{TF}} \) is the macroscopic ground-state energy from the Thomas–Fermi model of [340]. Neither quantity contains either shell or pairing effects. Note that there is no fit parameter in equation (6)! \( E_{gs-\text{nopair}}^{\exp} \) was taken from the 2012 Atomic Mass Evaluation [361, 362], averaged over odd–even staggering in \( Z \) and \( N \). The quantity \( E_{gs-\text{nopair}}^{\exp} - E_{gs}^{\text{TF}} \) defines the empirical ground-state shell correction, represented by \( \delta U_{gs} \) in figure 13.

In accordance with [359], figure 14 shows that the overall isotopic trend of the empirical barriers is very well reproduced by \( B_f^{\text{topo}} \). However, there are some systematic deviations in the absolute values for the different isotopic chains: Firstly, the barriers of thorium, protactinium and uranium isotopes are underestimated, while the barriers of the heaviest elements plutonium, americium and curium are underestimated. The deviation shows a continuous smooth trend as a function of \( Z \) with a kink at \( Z = 90 \). Secondly, a systematic odd–even staggering that is evident in the empirical barriers from protactinium to curium is not reproduced by the \( B_f^{\text{topo}} \) values estimated with equation (6).

In [355], an even better reproduction of the empirical barriers was obtained by applying a simple \( Z \)-dependent correction \( \Delta B_f \) to the values obtained by equation (6) and by increasing the pairing-gap parameter \( \Delta \) at the barrier in proton and neutron number to 14/√\( A \) MeV, which is appreciably larger than the average value of about 11/√\( A \) MeV found in the nuclear ground state [363]. This was technically done by increasing the binding energy at the saddle for nuclei with even \( N \) by 3/√\( A \) MeV and for even \( Z \) by the same amount. The correction term \( \Delta B_f \) was parametrized in a way to account for the observed \( Z \)-dependent deviations, as shown in figure 15.
The modified barriers are also shown in figure 14. Possible explanations for the origin of this correction are given in [228]. Note that the correction $\Delta B_f$ depends only on $Z$. Thus, it shifts the sequence of barriers of one element by the same amount and does not change the structures in these curves. Indications for an increased pairing gap at the barrier have already been discussed by Bjørnholm and Lynn [99] to explain the enhanced odd–even staggering at the saddle. They interpreted this finding as possible evidence for surface pairing.

In addition, figure 14 shows the predictions from $Z = 90$ to $Z = 96$ of an elaborate theoretical model [364] with a...
macroscopic–microscopic approach based on the meticulous mapping of the potential in five-dimensional deformation space [329]. Although as already mentioned, the fission barriers from this multi-dimensional model [364] cannot be directly identified with the one-dimensional fission barriers determined in [99], the comparison is not without interest. The model values [364] deviate appreciably from the empirical one-dimensional values [99] with a rms deviation of 1.42 MeV (see table 4). In particular, the isotopic trend is not well reproduced. Moreover, the model does not show the observed odd–even staggering of the barrier height. Other macroscopic–microscopic or microscopic models show similar deviations. In this context, it is interesting that nuclear ground-state masses can be obtained with an uncertainty of about 500 keV by the most reliable macroscopic–microscopic or microscopic models [365]. With this uncertainty and the additional uncertainty of the potential energy at the saddle in mind, uncertainties of the fission-barrier height from the macroscopic–microscopic approach on the order of 1 MeV are not unexpected. Thus, it seems to be doubtful whether the deviations between the empirical barriers [99] and those of [364] are related to the multi-dimensional character of the calculation. They may rather be an indication of the accuracy that the macroscopic–microscopic approach can obtain on the multi-dimensional potential-energy surface; see also the related discussion in [93].

The most interesting feature, however, is that the saddle-point masses (the nuclear binding energies at the saddle) extracted by Bjørnholm and Lynn [99], which are derived from energy-dependent fission cross sections and fission probabilities, show no structural effects beyond regular odd–even staggering. This is remarkable, because the underlying measured energy-dependent fission cross sections and probabilities, and (ii) the application of the topographic theorem for obtaining estimated values of the fission barriers are completely independent, the good agreement of these sets of values is a strong indication that, firstly, the empirical barriers deduced by Bjørnholm and Lynn represent the true fission thresholds in a one-dimensional picture with remarkable accuracy and that, secondly, the topographic theorem is a rather good approximation. From the rms deviation between these two sets of barriers, given in table 4, it may be concluded that the uncertainties of the empirical barriers, determined by Bjørnholm and Lynn are no larger than 500 keV, which is appreciably smaller than the presumed value of 1 MeV mentioned above.

The fact that the deviations can even substantially be reduced by a simple Z-dependent shift indicates that they are caused by systematic shortcomings in the Z dependence of the macroscopic model used for the estimations or by a slight violation of the topographic theorem, see [228] for a detailed discussion. By applying the deduced Z-dependent shift and increased odd–even staggering at the saddle, the empirical barriers are reproduced within their uncertainties of 200–300 keV, as given in [99]. Regarding the absence of any systematic deviations of the fission barriers estimated with the topographic theorem from the barriers determined by Bjørnholm and Lynn [99] along isotopic chains, it seems that it is well justified to assume that reliable predictions of fission thresholds in an extended region of the chart of the nuclides can be made with this description. The agreement of this parametrization with the empirical values proposed in RIPL 3 is less good (see figures 7 and 8 in [228]), in particular in the structures along isotopic chains, which are not affected by the applied Z-dependent shift. This gives more confidence to the empirical values of Bjørnholm and Lynn [99].

In a more fundamental sense, any noticeable structural effects on the fission-barrier height can be attributed to the microscopic contributions to the ground-state mass and to systematically stronger odd–even staggering at the barrier only. Any fluctuations of the saddle masses beyond even–odd staggering stay within the given uncertainties of the empirical fission thresholds [99], which typically amount to 200 keV. The influence of shell-correction energies on the saddle mass cannot strictly be excluded, but if there is any, it must show a gradual and smooth variation with Z and A, which behaves like a macroscopic quantity. At present, theoretical estimates of the fission barriers have not yet attained this accuracy; they

![Figure 15. The empirical correction applied to the fission-barrier height obtained with the topographic theorem as a function of the atomic number of the fissioning nucleus. Reproduced from [353]. © OECD 2014.](image-url)

| Table 4. The rms deviation between the empirical barriers and the values from different approaches. |
|-----------------------------------------------|----------------|---------|----------|----------|
| Topographic | Adjusted | Möller | RIPL 3 |
| 0.52 | 0.24 | 1.42 | 0.36 |

Note. The table lists the rms deviations in MeV of the different sets of fission barriers shown in Figure 14 and the values of RIPL 3 from the empirical values. References: empirical [99], topographic (this work, equation (6)), adjusted (this work and [355]), Möller et al. [364], RIPL 3 [103]. Note that [103] and [364] do not cover all nuclei, for which empirical values are available. The typical uncertainty of the empirical values, given in [99], is 0.2–0.3 MeV.
show deviations on the order of 1 MeV. The best reproduction of empirical fission barriers has been reported in [366], where a rms deviation of 0.5 MeV has been obtained within the framework of the macroscopic–microscopic approach. This is also the accuracy to be expected for theoretical calculations of the whole potential-energy landscape of the fissioning system.

4.3.3. Fission probabilities. The value of the highest barrier, deduced in the preceding section, is not sufficient to calculate the energy-dependent fission probability, which is required to estimate the probabilities of the different fission chances at higher excitation energies. In the scenario of a one-dimensional double-humped barrier with parabolic shapes around the saddles and around the second well, among others, the heights of both saddles and the curvature parameters are required. In [228], it was found from a fit to the empirical barriers of [99] that the quantity \( B_A - B_B \) in the actinides varies smoothly as a function of \( Z^3/A \) like

\[
\frac{(B_A - B_B)}{\text{MeV}} = 5.401 - 0.0066175 \cdot Z^3/A + 1.52531 \cdot 10^{-6} \cdot (Z^3/A)^2. \tag{7}
\]

Together with the estimation of the highest barrier described above and the systematics of other fission-barrier parameters, for example from [99], a rather accurate and complete set of fission-barrier parameters can be established for a region of nuclei, extending appreciably beyond the region covered by the experimental information. These parameters are used in GEF to provide the estimated fission probabilities in the framework of the transmission through, respectively, the passage over a one-dimensional, double-humped fission barrier. Additional ingredients are the nuclear level density, the gamma strength function and neutron transmission coefficients; for details we refer to [228].

Using the formalism of [99] assures a good reproduction of the measured fission probabilities and is expected to give rather accurate estimates for neighboring fissioning nuclei too. Further experimental information for specific fissioning systems is not required. However, this formalism does not provide any information on the resonance structures that appear at energies below and slightly above the fission barrier, because the appropriate parameters can only be deduced from dedicated experimental data, see section 4.4. Fortunately, these resonances have little influence on the characteristics of multi-chance fission due to the higher excitation energies that are mostly involved.

4.3.4. Multi-chance fission. When fission after particle emission is possible, the measured fission observables originate from the fission of several nuclides with different excitation energies. In this interpretation, the interpretation of the data and the modeling of the fission observables requires a good estimation of the characteristics of multi-chance fission. In GEF, the application of the topographic theorem and the scenario of the transmission, respectively passage, of a one-dimensional, double-humped barrier is used for this purpose. Thus, quantitative estimations of the contributions from different fissioning systems and the corresponding excitation-energy distributions

![Figure 16. The relative contributions of the different fission chances in the system \( {}^{235}\text{U}(n,f) \) as a function of the incident-neutron energy. Full line: first-chance fission, dashed line: second-chance fission, dot-dashed line: third-chance fission. Reprinted from [228], Copyright (2016), with permission from Elsevier.](image1)

![Figure 17. The distribution of excitation energies \( E^* \) at fission for the system \( {}^{235}\text{U}(n,f) \) at \( E_n = 14 \text{ MeV} \). \( E^* \) is the excitation energy of the compound nucleus above its ground state, before it passes the fission barrier towards scission. The right-most peak shows events from first-chance fission (fission of \( {}^{236}\text{U} \)), the middle curve corresponds to second-chance fission (fission of \( {}^{235}\text{U} \)), and the left curve corresponds to third-chance fission (fission of \( {}^{234}\text{U} \)). The broad distribution around 16 MeV corresponds to gamma emission before fission. Reprinted from [228], Copyright (2016), with permission from Elsevier.](image2)

at fission are provided. Figures 16 and 17 show the results for the system \( {}^{235}\text{U}(n,f) \).

4.3.5. Hidden regularities of fission channels. Although the good description of the fission barriers by the topographic theorem that is demonstrated in section 4.3.2 means that the saddle mass is essentially a macroscopic quantity, many other fission quantities show very strong signatures of nuclear structure, for example, the evolution of the shape and the potential on the fission path—in particular the existence of fission isomers, triaxiality at the first barrier and mass asymmetry at the second barrier due to shape-dependent shell effects in the actinides. Also, the fission-fragment yields are characterized by several components in the mass distributions from different fission channels that are attributed to shell effects in the potential energy and by an odd–even staggering in proton and
neutron number due to the influence of pairing correlations. The potential-energy surface of the $^{238}$U nucleus, calculated with the two-center shell model in [356], demonstrates the structures created by shell effects, see figure 18.

The mass-asymmetric deformation belongs to the relevant degrees of freedom of most dynamical fission models, and the manifestation of shell effects in the fission-fragment mass distributions plays a prominent role in benchmarking these models. Macroscopic–microscopic models, and to some extent microscopic self-consistent models as well, were rather successful in reproducing the appearance of mass-asymmetric fission in actinides [17, 299], the features of multi-modal fission around $^{258}$Fm [367], the gradual transition from single-humped to double-humped distributions around $^{226}$Th [32], and most recently, the appearance of complex mass distributions in a region around $Z \approx 80$ to $Z \approx 86$ from beta stability to the proton drip line [154]. However, the deviations from the measured data remain important, see the examples in figures 5–11. A much higher accuracy has been obtained with the semi-empirical description used in the GEF code [228] by exploiting regularities in the characteristics of the fission channels that are not obvious from the microscopic models, because these models treat each fissioning nucleus independently. In the following, we will describe the theoretical considerations that are behind this semi-empirical approach. They are not only important for the high-accuracy estimates of fission-fragment yields, kinetic energies, spectra and the multiplicities of prompt neutrons and gammas, as well as other fission quantities, but also for a better understanding of the fission process by revealing an astonishingly high degree of regularity in the properties of fission channels.

**Early manifestation of fragment shells.** When the two-center shell model [323] became available, it was possible to study the single-particle structure in a dinuclear potential with a necked-in shape. The investigations of Mosel and Schmitt [242, 368] revealed that the single-particle structure in the vicinity of the outer fission barrier indicates the existence of nearly independent single-particle states localized in the two fragments. This leads to a coherent superposition of the shell-correction energies from the two fragments. The authors explained this result by the general quantum-mechanical feature in which wave functions in a slightly necked-in potential are already essentially localized in the two parts of the system. Also, recent self-consistent calculations show this feature (e.g. [47, 62]), which is a direct consequence of the necking, independent of the specific shape parametrization. Therefore, one may expect that the complex structure of the fission modes can essentially be explained by the influence of the shells in the proton and neutron subsystems of the fragments. The potential-energy surfaces of fissioning systems calculated with the macroscopic–microscopic approach, for example [364], support this assumption. The fact that in actinides, where a double-humped fission-fragment mass distribution is observed, the theoretical models predict a mass-asymmetric shape at the outer saddle, also suggests that fragment shells are already established to a great extent at the outer saddle.

As a consequence, the shell effects on the fission path from the vicinity of the second barrier to scission can be approximately considered as the sum of the shell effects in the proton- and neutron-subsystems of the light and the heavy fission fragment. Thus, these shells do not primarily depend on the fissioning system but on the number of neutrons and protons in the two fission fragments. However, these shells may be substantially different from the shell effects of the fragments in their ground state, because the nascent fragments in the fissioning dinuclear system might be strongly deformed due to their mutual interaction [248].

Therefore, the potential energy can be understood as the sum of a macroscopic contribution, depending on the fissioning nucleus, which changes gradually on the fission path and from one system to another, and a microscopic contribution that depends essentially only on the number of protons and
neutrons in the nascent fragments. Thus, in nuclear fission, the macroscopic–microscopic approach turns out to be particularly powerful. The distinction of the two contributions to the potential is accompanied by an assignment of these contributions to different systems: the macroscopic potential is a property of the total system, while the shell effects are attributed to the two nascent fragments. The assumed well position at $Z = 55$ reflects the measured position of the asymmetric fission-fragment component (see figure 21). It is unclear whether this is an indication for a proton shell around $Z = 55$, see section 5.4. Reprinted from [228], Copyright (2016), with permission from Elsevier.

Quantum oscillators of normal modes. The early manifestation of fragment shells provides the explanation for the appearance of fission valleys in the theoretical calculations of the potential-energy landscape of fissioning nuclei, in particular in the actinides. As demonstrated in figure 18, these are valleys in the direction of elongation, starting in the vicinity of the second barrier until scission, with an almost constant position in the mass asymmetry. For the dynamic evolution of the fissioning system, this means that each valley can be considered as a quantum oscillator in the mass-asymmetry degree of freedom. The initial flux in each valley, corresponding to a specific fission channel, is decided at the outer barrier. Depending on the height of the ridge between neighboring fission valleys, there might be some exchange of flux further down on the way to scission. The positions of the asymmetric minima that are created by shell effects and the shape of the corresponding oscillator potentials stay approximately constant until scission, but the excitation energy of each quantum oscillator tends to increase on the way towards scission by feeding from the potential-energy gain along the fission path. It is assumed that the statistical ensemble of a large number of fission events establishes an excitation-energy distribution that can formally be replaced by the distribution of a quantum oscillator in instantaneous equilibrium with a heat bath of temperature $T$, whereby the temperature increases on the way to scission. With these ideas in mind, one can formulate the evolution of the mass-asymmetry degree of freedom of the fissioning system on the way to scission. Deviations from instantaneous equilibrium by a dynamical freeze-out will be discussed in the next section.

Stochastic calculations show that the excited nucleus stays in the ground-state minimum and later in the second minimum for quite a long time, see figure 4 in [35]. One expects that this is still true if tunneling is considered, because the transmission probability decreases by about five orders of magnitude per 1 MeV barrier height [372]. Therefore, an excited nucleus has enough time to re-arrange its available energy before passing the fission barrier. The probability for the passage of the fission barrier increases considerably, if the nucleus concentrates enough of its energy on the relevant shape degrees of freedom for avoiding tunneling as much as the available energy allows. If the available energy exceeds the barrier, this excess can be randomly distributed between the different states above the barrier without any further restriction, such that the barrier is passed with maximum possible entropy on average [373], again replacing event averaging with instantaneous equilibrium. For this reason, the fissioning system has no memory of the configurations before the barrier, except the quantities that are preserved due to general conservation laws: total energy, angular momentum and parity. Thus, the starting point for calculating the properties of the fission fragments is the configuration above the outer fission barrier.

Considering again the statistical ensemble of fissioning systems, the evolution of the entropy plays a decisive role in the fission process. The concentration of a large amount of energy into the elongation degree of freedom at the barrier

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10 As introduced by Gibbs [370], a statistical ensemble is an idealization consisting of a large number of virtual copies of a system, considered all at once, each of which represents the possible state that the real system might be in.

11 This assumption is supported by microscopic calculations reported in [77], which show that the dynamic evolution of the fissioning system beyond the fission barrier depends only a little on the initial configuration inside the first minimum chosen in the calculation.
leads to a decrease of the thermal energy and induces a reduction of the entropy\(^\text{12}\). After passing the barrier, the entropy may increase again due to dissipation. If dissipation is low, this increase may be small, and the passage from the barrier to scission may be adiabatic, leaving the entropy essentially unchanged. However, we think that the arguments for a reduction of the entropy on the way from the first minimum to the barrier, formulated above, are very strong. Therefore, we think that the approximation of treating fission as an isentropic process \([375, 376]\) is not a generally valid assumption.

Beyond the outer barrier, the distribution of the mass-asymmetry coordinate is given by the occupation probability of the states of the quantum oscillators in the respective fission valleys. The situation is schematically illustrated in figure 20 for the mass-asymmetry coordinate in two fission valleys that are well separated, assuming that the potential pockets have a parabolic shape. The fission-fragment mass distribution is given by the evolution of the respective collective variable, until the system reaches the scission configuration. It is defined by the number and the energy distribution of occupied states in the different valleys.

In the case of weak coupling and in thermal equilibrium with a heat bath of temperature \(T\), the ratio of the yields \(Y_i\) of the two fission channels corresponding to the population of the two harmonic quantum oscillators depicted in figure 20 is given by

\[
\frac{Y_2}{Y_1} = e^{-\Delta E/T} \cdot \frac{\hbar \omega_1}{\hbar \omega_2} \approx e^{-\Delta E/T}.
\]  

(8)

\(\Delta E = E_2 - E_1\) is the potential-energy difference between the lowest states in the two quantum oscillators, and \(\hbar \omega_i\) is the level spacing in the respective oscillator potential\(^{13}\). (Because the \(\hbar \omega\) values of the oscillators in the different fission values are normally rather similar, the factor \(\hbar \omega_1 / \hbar \omega_2\) in equation (8) is set to one as an approximation.) The distribution of the collective coordinate of the quantum oscillator for asymmetric distortions in one fission channel is a Gaussian function with a variance \(\sigma^2\) that is given by the well-known equation \([378]\)

\[
\sigma^2 = \frac{\hbar \omega}{2C \coth \left(\frac{\hbar \omega}{2T}\right)}.
\]  

(9)

\(C\) is the second derivative of the potential near its minimum in the direction of mass asymmetry.

If the exchange of flux between different fission channels beyond the second barrier is negligible, the temperature parameter in equation (8) is the value at the second barrier. As argued above, it is assumed to be given by the nuclear temperature at the second barrier as derived from the nuclear level density. However, the width of mass asymmetric distortions, described by the temperature parameter in equation (9), evolves on the way to scission. The width of the fission-fragment mass distribution is given by the temperature at the dynamical freeze-out, which is described in the following.

**Dynamical freeze-out.** It is well known \([379]\) that the statistical model, applied to the scission-point configuration, is unable to explain the variances of the mass and energy distributions and their dependence on the compound-nucleus fissility. Also, stochastic \([35, 328]\) and self-consistent models \([299]\) show the importance of dynamic effects on the width of the fission-fragment mass distributions, especially in low-energy fission. The studies of Adeev and Pashkevich \([380]\) suggest that dynamical effects due to the influence of inertia and dissipation on mass-asymmetric distortions can be approximated by considering the properties of the system at an earlier time. Nifenecker \([381]\) and Asghar \([341]\) explained the fluctuations in the charge polarization at scission by a freeze-out\(^{14}\) of the giant-dipole resonance before neck rupture. For all processes which are connected with the transport of nucelons from one nascent fragment to the other one (e.g. the evolution of mass asymmetry \([382]\) or charge polarization \([341, 381]\)), the effective mass increases dramatically during the necking-in of the

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\(^{12}\) We would like to stress that this reduction of entropy does not contradict the second law of thermodynamics, because the laws of thermodynamics have a statistical nature. Thermodynamical quantities, such as the entropy, are subject to fluctuations that become sizable in mesoscopic or microscopic systems like nuclei. A proper way to treat such systems is the explicit application of statistical mechanics \([374]\).

\(^{13}\) Equation (8) may be derived as follows: it is assumed that \(T > \hbar \omega\) and that the coupling between the oscillators and the heat bath is strong enough to allow for thermal equilibration; but it is so weak that the states of the oscillators are essentially undisturbed. In this situation, the population probability of a state \(n_i\) in oscillator 1 and the population probability of a state \(n_1\) in oscillator 2 are given by \(P_{1n_1} = \exp(-n_1 \cdot \hbar \omega_1 / T)\) and \(P_{2n_2} = \exp(-\Delta E - n_1 \cdot \hbar \omega_2 / T)\), respectively. The total population probabilities \(Y_1\) and \(Y_2\) in the two oscillators are obtained by summing up the individual population probabilities over \(n_i\) and \(n_1\), respectively:

\[
Y_1 = \sum_{n_1=1}^{\infty} P_{1n_1} \quad \text{and} \quad Y_2 = \sum_{n_1=1}^{\infty} P_{2n_2}.
\]

By replacing the discrete states with a continuous state density, the sums become integrals. This leads to the solution in equation (8).

\(^{14}\) The term ‘freeze-out’ in connection with fission-fragment yields was used by Asghar \([341]\) to denote the inability of the wave function of the fissioning system to adjust adiabatically when approaching scission.
fissioning system. This makes it more difficult for the system to adjust to the evolution of the potential-energy surface when approaching scission, which means that the statistical model may give reasonable results for the distribution of any observable related to a certain normal mode, if it is applied to a configuration that depends on the typical time constant of the collective coordinate considered. The relaxation time is specific to the collective degree of freedom considered. It is relatively long for mass-asymmetric distortions [383] and rather short for the charge-polarization degree of freedom [341, 381, 384, 385], due to the difference in the associated inertia. Thus, the shape of the potential and the value of the corresponding collective temperature, which are decisive for the distribution of the respective observable, are those that the system takes at the respective relaxation time before scission.

From these considerations, it may be concluded that the observed fission-fragment mass distribution of a specific fission channel can approximately be understood as the equilibrium distribution of the quantum oscillator in the mass-asymmetry degree of freedom in the corresponding fission valley at the time of freeze-out on the fission path with the local second derivative of the mass-asymmetric potential and temperature.

**Empirical extraction of universal fragment shells.** In the macroscopic–microscopic approach, the potential energies at the bottom of the different fission valleys, which determine their relative yields according to equation (8), are the sum of five terms: the macroscopic potential and the shell energies of the proton and neutron subsystems of the two nascent fragments. The stiffness of the macroscopic potential against mass-asymmetric distortions evolves gradually as a function of the fissility [314]. An empirical function has been deduced with a statistical approach [386] from the widths of the measured mass distributions of the symmetric fission channel at higher excitation energies, where the shell effects are essentially washed out. To describe the yields and the variances of the contribution of each fission channel to the fission-fragment mass distributions with equations (8) and (9), the following three parameters are required in addition to the curvature of the macroscopic potential: the position, the magnitude and the second derivative of each shell in each of the four subsystems (protons and neutrons in the light and the heavy pre-fragment). These parameters are not expected to depend on the fissioning system, and will stay constant on the way to scission, once the nascent fragments have acquired their individual properties.

Fragment shells deduced from the fission of actinides. It has been known for a long time that the mean mass of the heavy component in the asymmetric fission of the actinides is approximately constant at \( A \approx 140 \) [387]. This is an indication that the shells in the heavy fragment are dominant. Böckstiegel et al [273] compiled the systematics of the fission channels, deduced from the measured fission-fragment mass and element distributions, partly from the two-dimensional mass-TKE distributions. The systematics of the mean proton and neutron numbers of the standard 1 and standard 2 fission channels, following the nomenclature of Brosa et al [244], is shown in figure 21. Obviously, the standard 1 and the standard 2 channels are located at the proton numbers \( Z \approx 52 \) and \( Z \approx 55 \), respectively. This feature is most clearly evidenced by the data from the long isotopic chains measured in the electromagnetic fission of relativistic secondary beams [258], but it had already been observed for the proton-induced fission of isotopic chains of heavier elements by Gorodisskiy et al [388]. In contrast, the neutron number varies systematically as a function of the mass of the fissioning nucleus.

This means that the most prominent asymmetric fission channels, standard 1 and standard 2, are each caused by a fragment shell that fixes the number of protons in the heavy fragment at \( Z \approx 52 \) and \( Z \approx 55 \), respectively. However, while the value of \( Z = 52 \) is compatible with the \( Z = 50 \) proton shell, if some contribution from the neck is taken into account, one should be cautious to conclude that this observation is an indication for a neutron shell near \( Z = 55 \), or a bit lower due to the neck contribution. A discussion of this problem in view of the theoretical studies of the nuclear shell structure is found in section 5.4.1.

The shell strength behind the S2 fission channels was kept constant for all systems. The shell strength behind the S1 fission channel was assumed to vary as a unique function of the \( N/Z \) value of the fissioning system, starting from a maximum...
value for $N/Z = 82/50$—except for the plutonium isotopes, where this shell was enhanced by 0.4 MeV in order to fit the experimental mass distributions. This enhancement will come into play in the next section again.

The ideas outlined above, with a few refinements, were applied to describe the fission yields of a great number of fissioning systems ranging from $Z = 90$ to $Z = 112$ in the semi-empirical GEF model with remarkable accuracy, using a unique set of parameters [228]. Figure 22 shows the calculated fission-fragment pre-neutron mass distributions compared to the evaluated data for some selected nuclei. Note that the masses in the underlying data are unambiguously identified by radiochemical methods [260]. Thus, the GEF results can directly be compared with the evaluated data. This is not the case for the mass distributions determined in kinematical measurements (e.g. double-energy measurements), which are distorted by limited mass resolution [115] and ambiguities in the calibration [126], as already mentioned in section 3.2. In addition, the provisional pre-neutron masses of $^{260}$Md(sf) are shown in comparison with the GEF result. A much more detailed comparison covering many fissioning systems and different energies can be found in [228].

Fragment shells deduced from fissioning systems in the lead region. Let us recall that when moving from the actinides to lighter fissioning systems, asymmetric fission gradually

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Figure 22. The evaluated and measured mass distributions (black symbols) of fission fragments in comparison with the result of the GEF model (blue symbols). The mass distributions after prompt-neutron emission are taken from the evaluation of [389]. The provisional pre-neutron masses from the spontaneous fission of $^{260}$Md (black histogram) were directly deduced from the ratio of the fragment energies with a finite resolution of about four mass units without applying a correction for prompt-neutron emission. The data of the corresponding mass distribution are taken from [239]. The green lines show the calculated contributions from the different fission channels. Reproduced from [355] with kind permission of The European Physical Journal (EPJ).
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disappears, and symmetric fission takes over [258]. However, in the lead region, complex structures appear [252, 254], showing up as double-humped or triple-humped mass distributions [154]. It is tempting to search for regularities in this phenomenon, which would also allow us to improve the description of fissioning systems in this region with the GEF model. This appears to be a difficult task, however, because the data are very scarce.

Figures 23 and 24 show the measured mass distributions with the most pronounced structures in this region. For most of these distributions, the mean masses of the asymmetric components and their uncertainties were determined in [154].
Figures 25 and 26 show the deduced mean positions of these peaks in neutron and proton number, respectively, both in the light and in the heavy fragment as a function of the number \( N_{\text{CN}} \) of neutrons in the fissioning nucleus. The UCD assumption has been used to infer the number of protons and neutrons in the fragments from the measured mass distributions. Obviously, the positions in neutron number move in an irregular way. The same is true when the position in the proton number of the heavy fragment is considered. The position of the light peak in proton number, however, shifts much less and moves in a rather smooth way. Figure 26 also shows the position of the light fragment of the fission of Pu isotopes, belonging to the S1 fission channel, as shown.

Figures 25 and 26 show the deduced mean positions of these peaks in neutron and proton number, respectively, both in the light and in the heavy fragment as a function of the number \( N_{\text{CN}} \) of neutrons in the fissioning nucleus. The UCD assumption has been used to infer the number of protons and neutrons in the fragments from the measured mass distributions. Obviously, the positions in neutron number move in an irregular way. The same is true when the position in the proton number of the heavy fragment is considered. The position of the light peak in proton number, however, shifts much less and moves in a rather smooth way. Figure 26 also shows the position of the light fragment produced in the fission of plutonium isotopes, which corresponds to the S1 fission channel. As reported above, the yield of the S1 fission channel shows a clear enhancement when compared to the regular trend. One is tempted to attribute this enhancement to the simultaneous formation of heavy fragments near the doubly magic \( ^{132}\text{Sn} \) and the formation of light fragments in the fission valley, which is responsible for the double-humped peaks of fissioning systems in the lead region. The straight line shows a linear fit to the positions of the light peak in the fission of \(^{180}\text{Hg}\) and \(^{201}\text{Tl}\).

In this context, it is interesting to note that the mean \( Z \) values of the S1 and the S2 fission channels also show a modest linear variation as a function of mass \( A_{\text{CN}} \) or neutron number \( N_{\text{CN}} \), see figure 21. The magnitude of this variation is similar to the one observed in figure 26 for the shell around \( Z = 36 \) (around \( \Delta Z = 1 \) for \( \Delta N_{\text{CN}} = 10 \)), but the sign is the opposite. Maybe, the different sign is related to the fact that the shells behind the asymmetric fission channels in the actinides are located in the heavy fragment, while the structures in the measured mass distributions in the lead region are caused by a shell in the light fragment.

In order to reproduce the data, the strength of this shell was supposed to vary as a function of the number of neutrons in the fissioning nucleus \( N_{\text{CN}} \) as shown in figure 27. In addition, figure 27 shows the strength of the shell in the light fragment, which enhances the S1 fission channel in the Pu isotopes; see text for details.

Figures 23 and 24 show, in addition to the data, the description with the GEF\textsuperscript{15} code that was achieved after the implementation of a fission valley with the position defined by the straight line in figure 26.

In this context, it is interesting to note that the mean \( Z \) values of the S1 and the S2 fission channels also show a modest linear variation as a function of mass \( A_{\text{CN}} \) or neutron number \( N_{\text{CN}} \), see figure 21. The magnitude of this variation is similar to the one observed in figure 26 for the shell around \( Z = 36 \) (around \( \Delta Z = 1 \) for \( \Delta N_{\text{CN}} = 10 \)), but the sign is the opposite. Maybe, the different sign is related to the fact that the shells behind the asymmetric fission channels in the actinides are located in the heavy fragment, while the structures in the measured mass distributions in the lead region are caused by a shell in the light fragment.

In this case and in some others, where newly implemented features are involved, the calculations were made with the GEF version Y2017/V1.2, although this version is still under development, and the parameter values are still preliminary. This is indicated in the corresponding figure captions; in all other cases, version Y2016/V1.2 was used.
fissioning system, if one disregards the substantial deviations found between the GEF results and the measured mass distribution of $^{208}$Rn, see figure 23. Unfortunately, the statistical uncertainties for this system are rather large.

More data from future experiments will allow the tentative description of the structural effects in the fission-fragment distributions of nuclei in the lead region deduced in this section to be verified. In any case, this analysis has revealed some prominent systematic trends in the available data.

4.3.6. Heat transport between nascent fragments. The transformation of energy between potential energy, intrinsic and collective excitations as well as kinetic energy is a very important aspect of the nuclear-fission process. It determines the partition of the fission Q value (plus eventually the initial excitation energy of the fissioning system) between the kinetic and excitation energy of the final fragments. Moreover, the division of the total excitation energy between the fragments is of considerable interest, because it strongly influences the number of prompt neutrons emitted from the fragments. Thus, it also induces a shift towards less neutron-rich isotopes.

Dissipation on the fission path. As mentioned above, the description of dissipation in the fission process, in particular in low-energy fission, where pairing correlations play an important role, still poses severe problems to theory. In the range of pairing correlations, Bernard et al [15] developed the Schrödinger collective intrinsic model for describing the coupling between collective and intrinsic two-quasi-particle excitations on the fission path in an extended version of the generator coordinate method. Tanimura et al [62] recently observed deviations from the adiabatic limit of the microscopic transport theory by single-particle levels crossing in the vicinity of the fission barrier. Once the nascent fragments acquire their individual properties, the single-particle levels stay approximately parallel, and the process is essentially adiabatic. Shortly before scission, one-body dissipation becomes stronger due to the fast shape changes connected to the neck rupture.

The effects on the fission observables from these two processes are expected to be rather different. Because the relaxation time of intrinsic excitations is short compared to the estimated saddle-to-scission time$^{16}$, one may assume that the induced nucleonic excitations are, on average, equally distributed over all intrinsic degrees of freedom of the fissioning system when it reaches the scission configuration. The dissipation near scission, however, occurs so shortly before neck rupture that the equilibration of the energy may be expected to happen to a great part after scission, where the exchange between fragments is inhibited.

The dissipated energy is fed by the potential-energy difference between the outer barrier and scission for which Asghar and Hasse derived a general estimation [390]. This energy difference gives an upper limit of the dissipated energy, because it is shared by intrinsic excitations, collective excitations and pre-scission kinetic energy.

Statistical properties of the nascent fragments. As already mentioned, we assume the intrinsic excitation energy—consisting of the intrinsic excitation energy above the outer barrier plus the energy dissipated in the region of strong level crossing behind the barrier—to be, averaged over many fission events, equally distributed over all intrinsic degrees of freedom of the fissioning system when it reaches scission. The division of this excitation energy $E_{\text{tot}}$ among the nascent fragments in statistical equilibrium is determined by the number $N$ of states available in the two nuclei. Thus, the distribution of excitation energy $E_1$ of one fragment before neck rupture is calculated by the statistical weight of the number $N$ of states with a certain division of excitation energy between the fragments

$$
\frac{dN}{dE_1} \propto \rho_1(E_1) \cdot \rho_2(E_{\text{tot}} - E_1).
$$

Note that $\rho_1$ and $\rho_2$ are the level densities of the fragments in their shape just before scission, not in their ground-state shape! The remaining energy $E_{\text{tot}} - E_1$ is taken by the other fragment.

There exist several analytical level-density descriptions, e.g. [103, 391–394], and, recently, also a few microscopic calculations [395–397]. In the present context, we are not interested in describing the peculiarities of specific systems, but in understanding the main thermodynamical properties of a nucleus, which determine the average behavior of the energy division between the nascent fragments. For this purpose, using a global level-density description is better suited and more transparent than using the individual results of microscopic models for specific nuclei. General investigations of the validity of recommended parametrizations can be found in [103, 393, 398]. These were rather oriented in benchmarking the level-density descriptions against the empirical data derived from level counting, neutron resonances and evaporation spectra. However, in [399] it was pointed out that many level-density descriptions violate basic theoretical requirements, in particular in the low-energy range where pairing correlations play an important role. These violations are not often easily recognized or checked by a comparison with experimental data due to the incompleteness and uncertainty of the data, but they can be important when considering the evolution of nuclear properties in terms of statistical mechanics.

The requirements, formulated in [399], are:

- The level density below the critical pairing energy [400], the excitation energy where pairing correlations disappear$^{17}$, is characterized by an approximately exponential function, corresponding to a constant temperature. This is qualitatively explained by the phase transition from super-fluidity to a Fermi gas, which stores any additional energy in creating additional degrees of freedom by quasi-particle excitations instead of an increasing temperature.

$^{16}$ The characteristic time for the thermalization of the intrinsic excitation energy of a nucleus is a few times the time a nucleon needs to travel over the diameter of the nucleus with the Fermi velocity. This is on the order of a few times $10^{-22}$ s. The estimated saddle-to-scission time is appreciably longer, about $10^{-20}$ s [313] or even longer [68].

$^{17}$ Strictly speaking, this transition is not sharp due to the small size of the nucleus [403].
The resulting level-density description, proposed in [399], resembles the composite level-density formula of Gilbert and Cameron [391], however with an increased matching energy on the order of 10 MeV, see figure 1 of [399]. This value of the matching energy, which can also be interpreted as the critical pairing energy, is in good agreement with the results of an analysis of the measured angular distributions of fission fragments [410] and energy-dependent fission probabilities [392].

**Energy sorting.** From the previous discussion, we conclude that a fissioning nucleus on the way to scission develops from a mono-nucleus to a dinuclear system, where two nascent fragments acquire their individual thermodynamical properties well before scission. Because they are still connected by a neck, they can exchange nucleons and excitation energy [411]. At moderate excitation energies, the two nascent fragments form a rather peculiar system: they act like microscopic thermostats. Each fragment can be considered as a heat bath at a constant temperature for the other fragment, although the system has a rather small fixed number of particles and a rather low fixed amount of total energy. Disregarding shell effects, the nuclear temperature in the constant-temperature regime decreases systematically with the fragment mass: $T \propto A^{-2/3}$ [394].

Figure 28 illustrates the variation of the logarithmic slope of the level density of the fission fragments as a function of mass in the reduced energy scale. The nuclei are situated in the extreme light, respectively heavy, wings of the fission-fragment distributions in order to clearly show the effect. This indicates that the light fragment has a systematically higher temperature than the heavy one. According to an estimate on the basis of the empirical level-density description of von Egidy et al [394] (which considers the systematic variation of the temperature with the nuclear mass and the influence of shell effects) and using shell effects at scission from the fit parameters of GEF, the influence of shell effects may only inverse this tendency in nearly symmetric splits. This may eventually be the case for the S1 fission channel in the actinides. The few measured mass-dependent neutron multiplicities (e.g. [412, 413]) do not show any indication for such a reversed energy sorting, but, in any case, it would be difficult to explain.

---

18 The pairing condensation energy is defined as the energy difference between the (hypothetical) ground state without the pairing effect and that with the pairing effect [400].
to observe due to the small yield of the S1 fission channel in these systems.

As a consequence, the intrinsic energy (heat) tends to flow to the heavier nascent fragment that has a lower temperature. This process of energy sorting was first described in 2010 in [12]. The process of heat exchange proceeds in rather large and fluctuating steps, which lead to an averaging of the level densities and make possible the application of thermodynamical concepts [411]. This averaging significantly smoothes out the fluctuations in the level density present at the lowest excitation energies, which are caused by the first quasi-particle excitations. With increasing initial excitation energy, the fragments enter the Fermi-gas domain, and the energy sorting gradually disappears [21]. Asymptotically, at high excitation energy, the heat is shared by the fragments in proportion to their masses.

**Prompt-neutron yields.** There are several observables that provide information on the energetics of the fission process. The total kinetic energy of the fragments, and the energy spectra and multiplicities of prompt neutrons and prompt gammas are the most prominent ones. Among those, the prompt-neutron multiplicity gives the most direct and detailed information, because the emitted neutrons can individually be attributed to a specific fragment by their kinematical properties in a moving-source fit [414]. Moreover, neutron evaporation is by far the most probable decay channel, when the excitation energy exceeds the neutron separation energy. Thus, the excitation energy of a specific fragment is given to a good approximation by the sum of the neutron separation energies and the mean neutron kinetic energies, which can be estimated rather reliably, plus an offset, i.e. an amount of energy that ends up in prompt-gamma emission. This offset is rather independent of the initial excitation energy of the fragment. It amounts to about half the neutron separation energy of the final fission product plus the rotational energy that the fragment still has after prompt-neutron emission.

More than 40 years ago, the measurement of prompt-neutron multiplicities was already a subject of great interest, see for example [415–419]. Several experiments were performed to determine the mass-dependent average neutron multiplicity as a function of the initial excitation energy of the fissioning system. Figure 29 shows this kind of data for the neutron- and proton-induced fission of $^{237}$Np and $^{238}$U, respectively, for different excitation energies. It should be noted that the observed events from proton-induced fission are summed up from different fission chances. This means that the excitation-energy distribution at the saddle deformation reaches from the initial excitation energy down to energies in the vicinity of the fission barrier.

The curve for the fast-neutron-induced fission of $^{237}$Np in figure 29 shows saw-tooth behavior that is typical for the prompt-neutron multiplicities in actinides. An explanation in terms of the fragment shells that determine the deformation of the fragments at scission was given by Wilkins et al in their scission-point model [248]. As a result of their Strutinsky-type calculations, the energetically favorable deformation of the light and the heavy fragments increases with the mass of the fragment. This deformation energy is thermalized after scission and feeds the evaporation of prompt neutrons. The minimum around $A = 130$ is attributed to fragments near the doubly magic spherical $^{132}$Sn. Another salient feature of these data is that almost all additional energy induced by the increasing incoming-particle energy ends up in the heavy fragment. This feature remained unexplained for a long time in spite of many attempts. The discovery of energy sorting now provides a convincing explanation for the transport of essentially all additional excitation energy that is brought into the system to the heavy fragment. Similar features are found in the mass-dependent prompt-neutron multiplicities of the proton-induced fission of $^{243}$Pu with $E_p$ between 13 and

![Figure 29](image-url)

**Figure 29.** Post-scission mean prompt-neutron multiplicities as a function of fragment mass for different systems. The lines are drawn to guide the eye; the initial excitation energy of the fissioning system is listed. Reproduced from [420] with permission of The European Physical Journal (EPJ).

![Figure 30](image-url)

**Figure 30.** The measured prompt-neutron multiplicity in $^{237}$Np(n,f) as a function of the pre-neutron fragment mass at two different incident-neutron energies [413] (data points) in comparison with the result of the GEF model [228] (histograms). Reproduced from [353] © OECD 2014.
Odd-even effect in fission-fragment yields. The odd–even effect in fission-fragment distributions, i.e. the enhanced production of fission fragments with an even number of protons and/or neutrons, is one of the most prominent manifestations of nuclear structure. This phenomenon can be studied in analogy to the energy sorting, described above: Also with respect to pairing correlations, the individual fragment properties are assumed to be established well before scission [424], and statistical equilibrium before scission may be assumed.

As already mentioned, the nuclear level densities considered on an absolute energy scale evolve smoothly without any noticeable odd–even staggering as a function of the number of protons and neutrons—except the appearance of additional levels compared to odd–odd nuclei—with a fully paired configuration in even–even nuclei and with fully paired configurations in the proton, respectively neutron subsystem, in odd-A nuclei, see figure 28. Therefore, in a statistical consideration, the appearance of odd–even staggering in fission yields must be connected in some way with these fully paired configurations. Indeed, at reduced energies above the ground-state level of odd–odd nuclei, the statistical weight of excited states (see equation (10)) is equal in all classes of nuclei (even–even, even–odd, odd–even and odd–odd), if the smooth mass dependence is taken out. Also the number of available states in even–odd and odd–even nuclei above the ground-state level of odd-A nuclei is the same. This means that the overproduction of fragments with an even number of protons can be traced back to even–even light fragments that are formed fully paired at scission when statistical equilibrium is assumed.

A schematic model following these ideas was developed in [52]; see also [228] for the implementation in the GEF code. For an even–even fissioning nucleus, the number of configurations with an even number \( Z_1 \) of protons in one fragment, which implies that the charge of the complementary fragment \( Z_2 \) is also even, at a fixed total reduced energy \( U_{\text{tot}} \) is given by:

\[
N_{Z_1=\text{even}}^{\text{even}}(Z_1) = \int_{U_{\text{tot}}+\Delta_2}^{U_{\text{tot}}+\Delta_2} \rho_{1}(U_1)\rho_{2}(U_{\text{tot}} - U_1)\,dU_1
+ \int_{U_{\text{tot}}+\Delta_2}^{U_{\text{tot}}+\Delta_2} \rho_{1}(U_1)\rho_{2}(U_{\text{tot}} - U_1)\,dU_1
\]

where \( \rho_{1}(U_1) \) and \( \rho_{2}(U_{\text{tot}} - U_1) \) are the level densities of even–even and even–odd fragments, respectively. The mass numbers \( A_1 \) and \( A_2 \) of the two fragments (used to calculate the level density) are the closest integer numbers (even or odd for even–even or even–odd fragments, respectively) to the values calculated from \( Z_1 \) and \( Z_2 \) by the UCD assumption. The reduced energy \( U \) is shifted with respect to the excitation energy \( E \) available in the two nascent fragments \( U = E - n\Delta \), \( n = 0, 1, 2 \) for odd–odd, odd–mass, and even–even fragments, respectively. This ensures the use of a common energy scale in the frame of the fissioning system, which is a basic requirement for the application of statistical mechanics. Long-range variations of the total available excitation energy as a function of mass asymmetry [40] are neglected in the schematic model presented here. They might be additionally considered in a more refined model.

The number of configurations with an odd number \( Z_1 \) of protons in one fragment, which implies that \( Z_2 \) is also odd, for an even–even fissioning nucleus is:

\[
N_{Z_1=\text{odd}}^{\text{odd}}(Z_1) = \int_{U_{\text{tot}}+\Delta_2}^{U_{\text{tot}}+\Delta_2} \rho_{1}(U_1)\rho_{2}(U_{\text{tot}} - U_1)\,dU_1
\]
The local logarithmic four-point difference $\delta_p$ of the fission-fragment $Z$ distributions as a function of asymmetry, represented by the ratio of the nuclear charge of the light fragment $Z_1$ and the nuclear charge of the fissioning nucleus $Z_{CN}$. The symbols show the experimental data from the compilation of [275] and denote the target nuclei: $^{229}$Th (stars), $^{235}$U (open triangles), $^{242}$Cm (full triangles), $^{245}$Cm (open squares), $^{249}$Cf (open circles).

The lines correspond to the results of the model of [52] described in the text. Adapted from [52]. © IOP Publishing Ltd. All rights reserved.

$$+\int_{0}^{U_{\text{tot}}} \rho_1(U_1)(\rho_2(U_{\text{tot}} - U_1)) dU_1$$

where $\rho_1(U_1)(\rho_2(U_{\text{tot}} - U_1))$ are the level densities of odd–even and odd–odd nuclei, respectively, with the mass numbers $A_1$ or $A_2$. The mass numbers $A_1$ and $A_2$ of the two fragments are again related to $Z_1$ and $Z_2$ by the UCD assumption. The yield for even-$Z_1$ nuclei is $Y_{Z_1}(Z_1) = N_{\text{tot}}^{\text{ee}}(Z_1)/N_{\text{tot}}^{\text{oo}}(Z_1)$ with $N_{\text{tot}}^{\text{ee}}(Z_1) = N_{\text{ee}}^{\text{ee}}(Z_1) + N_{\text{ee}}^{\text{oo}}(Z_1)$. Similar equations hold for odd–even, even–odd and odd–odd fissioning systems. The total available reduced intrinsic excitation energy $U_{\text{tot}}$ is assumed to be a fraction of the potential-energy difference from saddle to scission, plus the initial excitation energy above the barrier [390]. Thus, it also increases with the Coulomb parameter $Z^2/A^{1/3}$ of the fissioning nucleus.

The result of these considerations is that the odd–even effect in the fission-fragment $Z$ distributions is caused by the statistical weight of configurations with a concentration of all intrinsic excitation energy and unpaired nucleons in the heavy fragment and the formation of a light fragment in a fully paired state.

This approach reproduces the observed salient features of the proton odd–even effect [275]: (i) the global odd–even effect $(\Sigma Y_{Z_{\text{ee}}} - \Sigma Y_{Z_{\text{oo}}}) / (\Sigma Y_{Z_{\text{ee}}} + \Sigma Y_{Z_{\text{oo}}})$ decreases with the Coulomb parameter and with increasing initial excitation energy. (ii) The local odd–even effect, represented by the logarithmic four-point differences, $\delta_p(Z + 3/2) = 1/8(-1)^{k+1}\ln Y(Z + 3) - \ln Y(Z) - 3[\ln Y(Z + 2) - \ln Y(Z + 1)]$, increases towards mass asymmetry. (iii) The local odd–even effect for odd-Z fissioning nuclei is zero at mass symmetry and approaches the value of even-Z nuclei for a large mass asymmetry. As shown in figure 31, the quantitative reproduction is satisfactory, except for the system $^{229}$Th($n$$_{th}$,$f$). The disagreement found for this system may have been caused by the neglect of fluctuations in the dissipated energy. In fact, for a great part of the fission events, the available energy of this system may be so low that they reach the scission point in a completely paired configuration due to the threshold character of the first quasi-particle excitation.

The same ideas are expected to be valid for the odd–even effect in fission-fragment $N$ distributions at scission, which means before the emission of prompt neutrons. However, in the measured number of neutrons in post-neutron fission fragments this initial odd–even effect is masked by the influence of the neutron-evaporation process, which imposes its own odd–even fluctuations [280, 425, 426]. This idea explains why the measured values of $\delta_p$ for electromagnetic and neutron-induced fission are very similar, see figure 1. It was successfully implemented in the SPACS code for calculating the nuclide yields of spallation residues [427].

We would like to stress that the model of [52] does not include the effect of the neck rupture. It is advocated by many authors (see [236]) that any production of odd-Z fragments starting from fully paired configurations at the saddle is exclusively caused by pair breaking during the fast shape changes connected with the rupture of the neck; however, no quantitative estimation has been proposed. This would imply that the motion from the saddle to the configuration before neck rupture is totally adiabatic and that a sizable fraction of the unpaired nucleons, emerging from the quasi-particle excitations at neck rupture, end up in different fragments. These assumptions can only be verified by elaborate microscopic models. The result of Tanimura et al [62] from TD-EDF theory seems to contradict the first assumption, because they obtained a sizable amount of dissipation before scission in the region of many local maxima across the viscosity of the second barrier. The second assumption is not obvious to us either, because we expect the localization of the wave functions in the dinuclear regime, discussed before, also to lead to a localization of the pairing correlations in the two nascent fragments already before scission. The later transfer of single nucleons from one nascent fragment to the other might be improbable during the short duration of the scission process. Finding a valid answer to these questions is an important task for dynamical quantum-mechanical models. In any case, the complete sorting of the available intrinsic excitation energy, consisting of the initial excitation energy of the system above the outer fission barrier and the energy that is dissipated between the outer barrier and scission, and its effect on the enhanced presence of an even number of protons and neutrons in the light pre-fragment, is expected to well describe the situation of the system before neck rupture, when the excitation energies of the fragments stay in the superfluid regime.

The odd–even effect in fission-fragment $Z$ distributions is one of the complex features of nuclear fission that can only be fully understood by dynamical quantum-mechanical models. These models need to be further developed by simultaneously handling dissipation, statistical mechanics and quantum localization in a realistic way. At present, the application of
statistical models [52] and considerations on the influence of dynamical processes at scission [236] give us an idea about the processes that are involved in the problem and that should be further studied.

4.4. Other results of the GEF model

While the underlying theoretical ideas of the GEF model were presented in section 4.3, together with the rather directly related observables, the present section deals with observables that are related to the basic ideas of GEF in a more complex way. The confrontation with the experimental data tests whether the interplay of the different elementary processes implemented in GEF is able to describe the complex behavior of the fission process in a realistic way in a more general sense. This section is not meant to be exhaustive. For additional information we refer to the detailed description of the GEF model in [228] and other specific publications, e.g. [428].

4.4.1. Fission-fragment yields at higher excitation energies. The fission-fragment mass distributions from the neutron-induced fission of $^{238}\text{U}$ for a series of incident-neutron energies [429] (black symbols) in comparison with the GEF-model results without accounting for (black dashed line) and accounting for (red full line) the experimental resolution. The neutron beam energy interval in the experiment as well as the energy used for the GEF calculations are indicated. Reprinted figure with permission from [428], Copyright (2018) by the American Physical Society.

However, for a full specification of the fission-fragment yields on the chart of the nuclides, the charge polarization (a different $N/Z$ ratio in the two fragments formed at scission) and prompt-neutron emission, which induces a shift to less neutron-rich fragments, need to be considered. (As already mentioned, the fission-fragment yields before prompt-neutron emission, fully defined in $A$ and $Z$, are practically inaccessible to experiment.) Data on the evolution of the $N/Z$ coordinate at higher excitation energies have emerged only recently from the VAMOS experiment. The measured average numbers of neutrons after prompt-neutron emission over $Z$ are shown in figure 33 and compared with the result of GEF calculations for different fissioning systems at different excitation energies. A pronounced structure above $Z = 45$ is observed, which is gradually washed away with the increasing mass and increasing excitation energy of the system. This structure is the combined effect of the charge polarization in the fission-fragment distribution before prompt-neutron emission, which favors the formation of neutron-rich heavy fragments in the $S_1$ and the $S_2$ fission channels, and the mass-dependent sawtooth shape of the prompt-neutron multiplicity. For a detailed discussion see, for example, [228]. The data are rather closely reproduced by the GEF model. Further investigations with the model suggest that the washing out of the structure can be attributed almost exclusively to the increasing excitation energy.

4.4.2. Total kinetic energy. Although the total kinetic energy (TKE) is given by the available energy of the system, which means by the sum of the initial excitation energy and the $Q$ value, minus the total excitation energy of the fragments, when the emission of particles and gamma radiation before scission is neglected, it contains additional information on the fission process. In particular, the mean TKE, averaged over all partitions in $Z$ and $A$ also reflects the changes of the $Q$ value due to variations of the fission-fragment distribution. TKE values measured after prompt-neutron emission also reflect the total corresponding mass loss of the system before and after the fission process.

Figure 32. Measured fission-fragment mass distributions from the neutron-induced fission of $^{238}\text{U}$ for a series of incident-neutron energies [429] (black symbols) in comparison with the GEF-model results without accounting for (black dashed line) and accounting for (red full line) the experimental resolution. The neutron beam energy interval in the experiment as well as the energy used for the GEF calculations are indicated. Reprinted figure with permission from [428], Copyright (2018) by the American Physical Society.

Figure 33. The mean post-neutron fragment $N/Z$ ratio as a function of the fragment $Z$ for various fissioning systems as measured at VAMOS (symbols) [430], and compared with GEF calculations (dashed lines). Reprinted figure with permission from [428], Copyright (2018) by the American Physical Society.
scission. Thus, model predictions on the fragment yields can also be tested indirectly by experiments other than direct measurements of the yields. This also shows again the interconnections and the correlations between the different fission quantities.

Figure 34 shows the recently measured average TKE values from the neutron-induced fission of $^{235}\text{U}$ [431, 432] in comparison with the GEF code. These data are fairly well reproduced, although some deviations above $E_n = 40 \text{ MeV}$ appear, whose origin is not clear. Detailed investigations also show that the total prompt-neutron yields are well reproduced (they have been measured up to $E_n = 50 \text{ MeV}$ [433, 434]), and lead to a reduction of the fragment masses and, thus, explain part of the TKE decrease at higher energies. Good agreement with the measured mass distributions of $^{238}\text{U}(n,f)$ [429], demonstrated in figure 32, indicates that the fission-fragment mass distribution, which determines the average $Q$ value, is also well reproduced.

The kink of the calculated values in figure 34 at about $E_n = 14 \text{ MeV}$ is related to multi-chance fission. Here, third-chance fission, which means fission of the compound nucleus $^{234}\text{U}$, sets in. This leads to a reduced yield of symmetric fission (with systematically lower TKE), and an enhanced yield of asymmetric fission (with systematically higher TKE). The nucleus $^{234}\text{U}$ has a large fission probability at an energy range somewhat above the fission barrier, because the fission barrier $B_f$ is lower than the neutron separation energy $S_n$. This effect is weaker at the onset of second-chance fission because in $^{235}\text{U}$ we have $B_f > S_n$. This kind of structure disappears at higher energies, because it is washed out by the fluctuations of the kinetic energies of the pre-fission neutrons.

4.4.3. Emission of prompt neutrons and gammas.

Pre-scission neutrons. There are at least two sources of neutrons emitted before scission. The first pre-scission neutron source is caused by the competition of particle emission and fission of the initial excited nucleus in the first well, eventually under the influence of transient effects [435]. Transient effects delay the onset of the quasistationary probability flow over the fission barrier under the influence of dissipation after the formation of the fissioning system. If the initial excitation energy is high enough, fission can occur from the initial nucleus, and after the emission of mostly neutrons from the respective daughter nuclei. This phenomenon, called multi-chance fission, has an influence on the fission probabilities and the fission-fragment properties, as already discussed in section 4.3.4, but it also has an influence on the energy spectra, the angular distribution, and the multiplicity of the prompt neutrons. The second pre-scission neutron source is fed by the evaporation of neutrons on the fission path, essentially beyond the outer fission saddle. This phenomenon has been intensively investigated to determine the nuclear dissipation strength [420]. In an experiment, neutron emission before scission can be identified by a kinematical multi-source fit [436], but the emission in the first well and the one beyond the fission barrier cannot be distinguished by this method.

The dependency of the neutron emission between the saddle and scission on the properties of the fissioning systems is complex and not fully understood, see e.g. [437]. Therefore, in the GEF code a heuristic approach is used. Guided by the saturation of the total multiplicity of the neutrons emitted from the fragments at about four neutrons found in [438], saddle-to-scission neutrons are assumed to be emitted as long as the excitation energy of the fissioning nucleus at scission exceeds 40 MeV.
Post-scission neutrons. After neck rupture, the fragments still interact with the Coulomb force, which leads to an increase of the relative velocities, until they reach their final kinetic energies. Only for a very short time, when the gradient of the force across the fragments is still large, may additional interactions also occur, like Coulomb excitation and the generation of collective fragment angular momentum \([439]\). After this, the fragments can be considered separately as independent nuclei, carrying intrinsic and collective excitations as well as a well-defined angular momentum. The fragment shape at scission that is influenced by the interaction with the complementary fragment relaxes to the equilibrium shape, and the gain in binding energy by the smaller deformation energy thermalizes and adds up to the initial intrinsic excitation energy. Also, the excitation energy stored in collective excitations transforms into intrinsic excitations, while preserving the fragment angular momentum.

Thus, the fragments after scission can essentially be considered as compound nuclei with well-defined excitation energy and angular momentum. They will undergo a statistical de-excitation process that can be described by a standard evaporation code. At sufficiently high excitation energies, some particle emission during the acceleration phase may need to be considered, but most of this process occurs from the fully accelerated fragments.

The characteristics of the emitted particles (mostly neutrons) and of the gamma radiation are important for two reasons: firstly, they carry precious information on the evolution of the fissioning system before scission; secondly, they are important for the design and operation of nuclear-power reactors.

Therefore, a statistical-model code is implemented in the GEF code that calculates the prompt neutrons and the prompt-gamma radiation emitted from the fragments after scission. The kinematical properties of the neutrons refer to the velocity of the emitting source. Isotropic emission in the respective frame of the moving fragment is assumed. The angular momentum is assumed to be conserved during the de-excitation process, because changes are expected to be small due to the low angular momentum values involved, where the influence of the angular-momentum-dependent level density and of the slope of the yrast line tend to compensate each other \([440]\); for details, see \([228]\).

Neutron multiplicities. Direct measurements of post-scission prompt-neutron multiplicities are difficult and subject to sizable uncertainties. Very precise values have indirectly been

Figure 36. Upper panels: experimental prompt-fission-neutron energy spectra (black lines and error bars) for \(^{235}\text{U}(n_{th},f)\) \([443]\) (left part) and \(^{252}\text{Cf}(sf)\) \([444]\) (right part) in comparison with the result of the GEF model (red histograms) on a logarithmic scale. In the lower panels, the spectra have been normalized to a Maxwellian with \(T = 1.32\) MeV and \(T = 1.42\) MeV, respectively. Reproduced from \([442]\). CC BY 4.0.
deduced from integral and other kind of experiments; however, they may deviate from the true values, because they are tuned together with other not accurately known fission quantities to exactly reproduce the results of the indirect experiments. Therefore, the results from direct experiments on prompt-neutron multiplicities in the spontaneous fission (sf) of several systems are compared with the results of the GEF code in figure 35. The data are reproduced with a standard deviation of 0.1 units. One can observe the sharp drop of the calculated prompt-neutron multiplicities in the transition from 256Fm(sf) to 258Fm(sf). Also, the peculiar slope of the prompt-neutron multiplicities as a function of mass in the Pu systems, which differs from the average trend, is a structural effect due to the strong and strongly varying yield of the S1 fission channel in these systems. These features demonstrate the importance of these data for the understanding of the fission process.

More results on the characteristics of prompt-neutron multiplicities are shown in [228].

Neutron-energy spectra. In figure 36, the energy spectra of prompt post-scission neutrons are compared with the experimental data for 235U(nth,f) and 252Cf(sf). The GEF code establishes a strong correlation between the shape of the spectrum (in particular its hardness) and the prompt-neutron multiplicity. The high-energy tail is sensitive to the yields and the deformation of the heaviest fragments of the light group, because they have the highest nuclear temperatures. The model parameters were slightly tuned to simultaneously describe the measured mass yields, the mass-dependent prompt-neutron multiplicities and the high-energy tails of the spectra for 235U(nth,f) and 252Cf(sf) shown in figure 36, without deteriorating the agreement with other fission observables. Due to the rather good agreement of the fragment properties at scission with the measured data provided by the GEF model, calculations with these parameters are also expected to give realistic predictions for prompt-neutron spectra and multiplicities for other systems in this region, including for those where no experimental data are available.

Prompt-gamma emission. As a weak decay channel, gamma radiation is emitted in competition with neutron emission and fission from the excited nuclei from the beginning on, while it becomes the dominant and finally the only decay channel when the excitation energy falls below the neutron-separation energy and the fission barrier. The same is true for gamma radiation emitted from the fragments, although here the fission branch is negligible. Gamma radiation observed in coincidence with fission is largely dominated by emission from the fragments after scission. Non-statistical gammas in coincidence with fission are exclusively observed from the fragments.

Statistical gammas. At energies sufficiently high above the yrast line, E1 radiation is dominant. It is essentially continuous and mostly governed by the gamma strength of the giant dipole resonance.

Figure 37. The low-energy part of the experimental gamma-energy spectrum (black line with error bars) [161] for thermal-neutron-induced fission of 235U in comparison with the GEF prediction (red full line). The GEF result is convoluted with the experimental energy resolution. In addition, the calculated spectrum, which includes the known delayed isomeric gamma transitions reported in JEF 3.1.1, is shown by the light blue line. Reprinted from [228], Copyright (2016), with permission from Elsevier.

Non-statistical gammas. At energies close to the yrast line, distinct gamma lines appear, and, finally, E2 radiation takes over, most efficiently removing the fragment angular momentum. The transition energies are specific to the nucleus considered, and accurate theoretical modeling is very demanding, e.g. [445]. Therefore, if possible, tables of experimental spectroscopic data or, eventually, empirical descriptions are often used, like the variable-momentum (VMI) model [446]. Compared to other advanced versions of the VMI model, e.g. [447], the implementation of the VMI model in GEF [228] provides improved modeling of the transition from rotational to vibrational behavior near closed shells, see [228].

The low-energy part of the measured prompt-gamma spectrum of 235U(nth,f) is compared with the result of the GEF code in figure 37. In this calculation, only the E2 gamma cascade along the yrast line is included, until the fragment meets the angular momentum of a known isomeric yrast state, which is listed in the JEFF 3.1.1 decay library. The measured spectrum is fairly well reproduced with the exception of a structure around 0.9 MeV, which is overestimated by GEF. Many gamma lines in this region stem from nuclei close to the N = 82 or the Z = 50 closed shell, where many isomeric transitions have been observed. This is illustrated by the blue line, which includes the full E2 gamma cascades below the known isomeric states contained in the JEFF 3.1.1 decay tables. A possible explanation for the over-estimation by the first-mentioned GEF calculation could be that it still contains some isomeric transitions, which are not listed in JEFF 3.1.1, because they are too short-lived or are not yet known and do not fall into the detection time window of the experiment.
4.4.4. Delayed processes. Due to the curvature of the beta-stability line on the chart of the nuclides, the fission fragments are in general unstable against $\beta^-$ radioactive decay. $\beta$ decay is a slow process with half lives in the ms region or longer. Thus, this process is well separated in time from the emission of prompt neutrons and gammas; the beta-decay chain ends when a stable nucleus is reached.

**Beta decay.** Beta decay proceeds to excited levels or to the ground state of the daughter nucleus with different probabilities, given by the beta-strength function. The available energy (the $Q$ value minus the rest energies of the electron and anti-neutrino) is shared between the emerging electron and the accompanying anti-neutrino [448].

**Delayed gamma emission.** If the beta decay populates an excited level in the daughter nucleus, this nucleus decays in most cases by one or more gamma transitions to their respective ground state, eventually further delayed by an isomeric state.

**Delayed-neutron emission.** When the beta $Q$ value of very neutron-rich fission fragments exceeds the neutron separation energy, the emission of delayed neutrons opens up as a possible decay channel. The delayed neutrons play a major role in reactor control, and many coordinated efforts have been undertaken to explain the complex characteristics, in particular in the excitation-energy dependence, of the delayed-neutron yields [449–451]; however, this problem has not yet been solved [452].

The GEF code follows the radioactive-decay chains, calculates the cumulative yields [453] and provides a list of delayed-neutron emitters with their contributions to the delayed-neutron yields. The decay tables of JEFF 3.1.1 are used. Figure 38 shows the calculated delayed-neutron yields in comparison with the experimental data for the neutron-induced fission of different systems. The calculated curves show some systematic features:

(i) There is a more or less pronounced plateau at low energies up to about 4 MeV. This plateau is caused by the combined effect of increased prompt-neutron emission, which tends to decrease the yields of the most neutron-rich fragments, and the gradual decrease of the odd–even staggering in the $Z$ yields, which tends to enhance the yields of odd-$Z$ fragments. Note that delayed neutrons are predominantly emitted from isotopes of odd-$Z$ elements, which have a systematically higher beta $Q$ value.

Figure 38. The probability for the emission of delayed neutrons for the neutron-induced fission of different systems as a function of the incident-neutron energy. The experimental data (full black symbols) from experimental nuclear reaction data (EXFOR) [453] are compared with the result of the GEF code (version Y2017/V1.2) (open red symbols, connected by red lines.)

Figure 39. The global odd–even effect ($\Sigma(Y_{\text{even}} - Y_{\text{odd}})/\Sigma(Y_{\text{even}} + Y_{\text{odd}})$) in the fission-fragment $Z$ distribution of the system $^{239}$Pu(n,f) as a function of the incident neutron energy from the GEF code.

---

19 The cumulative yield $Y(A,Z)$ of the nuclide $(A,Z)$ is the total number of atoms that the nuclide produced over all time after one fission.
Both effects tend to compensate each other. The plateau disappears when the odd–even staggering in the $Z$ yields is switched off in the calculation. The plateau is most pronounced for $^{232}$Th(n,f), which shows the largest odd–even staggering in the fission-fragment $Z$ yields (see figure 31). It is absent in odd-$Z$ fissioning systems, for example in $^{241}$Am, because the odd–even staggering in the $Z$ yields is much smaller, except at large mass asymmetry [275].

(ii) Above this plateau, there is a fall-off, which is most clearly seen in the case of even-$N$ target nuclei. Often, this fall-off is attributed to the loss of one neutron at the onset of second-chance fission [454, 455]. The mechanism becomes clear when the energy-dependence of the odd–even effect in the fission-fragment $Z$ distribution is considered, which is shown in figure 39. When second-chance fission sets in, the odd–even staggering in the fission-fragment $Z$ distribution shows a peak. This effect is especially strong for even-$N$ target nuclei due to the high fission probability slightly above the fission barrier of the even-$N$ isotope, which is formed after the emission of one neutron in the case of an even-$N$ target nucleus. This reduces the relative yields of the odd elements, which are predominantly responsible for the delayed-neutron production.

(iii) Above about 6–7 MeV, there is a gradual decrease of the delayed-neutron yield with the increasing energy of the incident neutron in all systems. This gradual decrease is understood by the shift of the isotopic distributions towards less neutron-rich isotopes by the increasing emission of prompt neutrons. This is again partly compensated by the gradual reduction of the odd–even staggering in the contribution from second-chance fission, in a similar manner.
way as in the first plateau, thus reducing the slope of the energy-dependent delayed-neutron yield in this region. (iv) A bump appears in some cases near $E_n = 15$ MeV, slightly above the threshold of third-chance fission. This structure may be connected with the high fission probability slightly above the fission barrier of the even-$N$ isotope, which is formed after the emission of two neutrons in the cases of an odd-$N$ target nucleus, although the mechanism is not clear.

From the comparison with the measured data, one can deduce that the strong variation of the delayed-neutron yield from system to system is fairly well reproduced by the GEF code. The first plateau and the fall-off show up rather clearly in the measured delayed-neutron yields of all uranium isotopes and of $^{232}$Th. For $^{238}$U and $^{232}$Th, the GEF results also show a clear fall-off, although it is not as pronounced as in the data. Some indications for a fall-off are also seen in the GEF results for $^{235}$U and $^{233}$U. The experimental data are too scarce, and they scatter too much, to pin down the slope of the gradual descent above the fall-off, but it seems that it is somewhat overestimated by GEF. The measured data are also too scarce to verify the predicted bump at the onset of third-chance fission, maybe with the exception of $^{232}$Th, where, however, this bump does not appear in the calculation.

A much more detailed documentation of the GEF calculations is shown in figures 40 and 41, where the contributions of the delayed-neutron emitters in the light and the heavy fission-fragment group to the delayed neutron yield are shown on a chart of the nuclides for the system $^{238}$U(n,f). Evidently, the neutron yields in the light group stay almost constant, in contrast to the yields in the heavy group, which decrease strongly with increasing excitation energy. This different behavior is certainly caused in great part by the energy sorting, discussed in section 4.3.6, which assures that the isotopic distributions in the light fission-fragment group do not move when the excitation energy varies. Only at the onset of second- (or higher-chance) fission is a slight shift expected, but obviously it does not have a noticeable effect on the delayed-neutron yields. Figure 41 also reveals a strong reduction of the contributions from even-$Z$ fragments to the delayed neutron yields with increasing incident energy, which is most clearly seen in the heavy fission-fragment group. This confirms the important role of the odd–even effect in producing the plateau in the delayed-neutron yields below 4 MeV.

It seems that the GEF calculations come rather close to understanding the origin of the observed structural features in the energy-dependent delayed-neutron yields, although some quantitative deviations show up. Two effects can clearly be identified, which strongly determine the evolution of the delayed-neutron yields with increasing excitation energy in the model: the odd–even staggering in the fission-fragment yields and the shift of the fission fragments towards less neutron-rich isotopes by prompt-neutron emission; and also the growing yield of the symmetric fission channel as well as other changes in the fission-fragment yields certainly have some influence. It is noteworthy that the cases in which the sequence ‘plateau, fall-off, gradual decrease’ is least pronounced in the GEF results, are those with the lower total delayed-neutron yields. In these cases, the neutrons are provided by nuclei with low delayed-neutron branchings. This gives rise to the suspicion that the uncertainties in the decay data may also play a role.

At the moment, the reason for the observed deviations is not clear. It is also not that easy to adapt the model in some way, because there are constraints from other observables. For example, the measured energy-dependent prompt-neutron multiplicities (see figure 77 in [228]), TKE values (see figure 34) and fission-fragment yield distributions (see figure 33) are rather well reproduced by the model. Still, there are no sufficiently accurate data available to pin down the shape of the function that describes the excitation-energy dependence of the odd–even staggering in the fission-fragment Z distribution [503], due to the inability to vary the excitation energy precisely in a well-defined way.

Already in previous theoretical considerations, it was mostly the prompt-neutron emission [456, 457] and the odd–even fluctuations in the fission-fragment yields [458] that were connected with the observed features of the delayed-neutron yields. In GEF, all the effects of the comprehensive description of the fission process are included in a consistent way, as much as they are known. The GEF model is able to reproduce the prominent characteristics of delayed-neutron probabilities to a certain extent and connect them with certain features of the yields and the excitation energies of the fission fragments—although the delayed-neutron data have not yet been exploited during its development. There is good agreement in the absolute values and their variation from system to system, while the structures are at least qualitatively reproduced. Thus, the complex features observed in the energy dependence of the delayed-neutron yields seem to be essentially understood. The key is the general coverage of practically all fission observables by the GEF model. GEF is certainly also well suited to the development of a more accurate quantitative description of the delayed-neutron yield, which is consistent with all other relevant observations, when the delayed-neutron data are included in the further development of the model.

**Decay heat.** Decay heat is the energy released by beta decay, and the $\beta$-delayed neutrons and gammas of the fission products. It is the only source of heat in the nuclear fuel rods after the reactor shutdown [459]. Knowledge of the amount and form of energy emitted in the radioactive decays of fission products is critical for the determination of safety procedures for nuclear power plant operation and for the cooling of nuclear fuel after an accidental or planned reactor shutdown.

The calculated decay heat following a thermal fission burst of $^{235}$U is shown in figure 42, using different evaluated fission-fragment yields, and compared with the experimental data. Apparently, the values based on the GEF yields are rather close to the values based on the evaluations. The parameters of GEF2017/1.2 were adjusted to the evaluated fission yields from ENDF/B-VII. No specific adjustment to the measured decay-heat data was performed.

**4.4.5. Correlations.** The GEF model is unique in providing a global description of the complete fission process, in which
all features are described in a consistent way and linked to each other with all their interdependencies in full detail. This is an important asset for advancing the understanding of the fission process. It also entails that the GEF model has the potential to revolutionize the application of nuclear data for technology by replacing completely separate or insufficiently linked descriptions of the specific features, which are adjusted independently to a small subset of observables and which are often incompatible with each other. In particular, the presently used mostly technically defined covariances or correlations between specific fission observables [460–462] are supplemented by correlations, which are defined by the physics of the fission process, between any pair of fission quantities. Correlations are also provided between the fission observables of different fissioning systems.

4.5. Other models

We have seen in the previous sections that the semi-empirical GEF model covers almost all fission quantities, starting from the fission probabilities, multi-chance fission, pre-scission neutron emission, and fission-fragment isotopic yields and kinetic energies, to the statistical de-excitation of the fission fragments, and, finally, to the delayed processes after beta decay with all their correlations and interdependencies. This assures that the different fission quantities are consistent, which can have an important impact on the results. The self-consistent and the stochastic models are much more restricted, but they are indispensable for exploring specific fundamental problems, for example, about the fission times or the shell effects in the fission observables of systems in unexplored regions. One may state that GEF covers all fission phenomena that can be ‘generalized’: if one knows the connected quantities for a certain number of systems, one can conclude on their values for other systems without the need for additional specific experimental information. As an example, macroscopic nuclear quantities are usually slowly varying, when the composition in terms of the neutron and proton number of the system gradually changes. The same is true as a function of excitation energy and angular momentum. Thus, these quantities can be generalized, which means that they can be reasonably well interpolated, and, to some extent even extrapolated if sufficient empirical information is available. Also, microscopic (e.g. shell) effects beyond the outer saddle can be generalized, because they can be understood as the superposition of microscopic effects in the neutron- and proton-subsystems of the fissioning nucleus due to the early localization of the wave functions [242] in the nascent fragments. This generalization extends over all systems that produce fission fragments in a region in $N$ and $Z$, for which information on these microscopic effects (e.g. the depth and shape of fission valleys, the deformation of the nascent fission fragments at scission) can be deduced from experimentally explored fission-fragment properties.

However, this method has some limitations. In particular, the resonances in the fission cross section induced by low-energy neutrons and the sub-barrier structure due to the coupling between states in the first and the second well of the potential as a function of elongation cannot be generalized due to their complexity. GEF can only provide a smooth average result. Also, for the accurate calculation of the prompt-gamma spectrum—in particular the contribution from non-statistical gammas—the exact experimental knowledge of the spectroscopy of the fission fragments is required. In this case, GEF offers an approximate solution with its improved VMI (variable moment of inertia) model, which, however, does also have the advantage of providing rather good results for cases where spectroscopic data are missing. In the following, some dedicated models for these problems will be described, which may be applied if the relevant experimental information is available. Their main importance lies in the application to nuclear-reactor technology.

4.5.1. Fission cross section. The nuclear fission cross section is highly fluctuating and finely structured. At energies below the fission threshold, resonances appear due to the coupling between states in the first and second, and eventually, in the third minimum. In neutron-induced reactions, which are most important for nuclear-reactor technology, neutron resonances in the neutron-capture cross section reflect the levels in the compound system. They create a dense pattern of resonances, which are separated up to a certain excitation energy (in the resolved-resonance region). They also influence the fission cross section in fissile nuclei, in which the fission barrier is lower than the neutron separation energy. At higher energies, in the unresolved-resonance region, one still observes fluctuations. Only at even higher energies, in the continuum region, where the levels overlap, does the capture cross section vary smoothly as a function of neutron energy. Moreover,
the transition states above the saddle points create a pattern in the fission cross section at energies slightly above the saddle energies. Structures in the fission cross sections appear again around the onset of the first- and higher-chance fission, due to the fissioning nuclei that proceed to fission at energies close to their fission barrier.

Much effort has been invested in an accurate description of the nuclear fission cross section with all its complexity [99–109]. In any case, these models rely on a tremendous number of parameters that have to be deduced from the experimental data, to a great part for each single system independently. These are very accurate parameters, which define (i) the heights of the fission saddles, the depths of the minima and the corresponding potential curvatures to deduce the transmission, considering the energies of the states in the second (and eventually also in the third) minimum, (ii) the parameters of the R-matrix theory [463], which define the resonance energies, and (iii) the energies and other properties of the transition states above the saddle points, as well as several other parameters. Several model codes contain very elaborate model descriptions that are able to reproduce complex data very well, for example the GNASH code [464], the CCONE code [465], the EMPIRE code [466], the CHO code [467], the TALYS code [468] and the CONRAD code [469]. Some of the recent developments for the description of the fission cross section include improved phenomenological optical model potentials for neutrons and protons with incident energies from 1 keV up to 200 MeV [470], the development of a model for the transmission through a triple-humped fission barrier with absorption [466], the parametrization of the threshold behavior near the fission barrier by an effective barrier distribution, the extraction of an effective purely experimental fission barrier, a heuristic model of resonant transmission in [108], and the extraction of one-dimensional fission barriers from potential-energy surfaces calculated with macroscopic–microscopic models to obtain fission transmission coefficients that can be used in a Hauser–Feshbach model in [469].

4.5.2. Fragment de-excitation. Most of the excitation energy of the fission fragments is liberated by the evaporation of light particles. Because the fragments emerging from the fission of nuclei close to beta-stability are very neutron-rich with respect to beta stability, these are almost exclusively neutrons. The energy spectra of these neutrons, as well as their multiplicity, belong to the most important nuclear data for the design and the operation of nuclear-fission reactors. The modeling of the energy spectra, and to some extent the modeling of prompt-neutron multiplicities as well, starting from empirically deduced information on the excitation-energies of the fragments and some additional ad hoc assumptions, e.g. about the division of the excitation energy between the two fragments, has a long tradition.

One of the first widespread descriptions of the prompt-neutron spectrum was introduced by Watt [471]. He proposed a closed formula, deduced from a Maxwell-type energy spectrum from one or two average fragments and the transformation into the frame of the fissioning system with at least two adjustable parameters: the temperature and the velocity of the average fragment. The Los Alamos model [330] extended this approach essentially by using a triangular temperature distribution of the fragments to a four-term closed expression for an average light and an average heavy fragment. A similar two-fragment model was also used by Kornilov et al in [472]. In 1989, Madland et al [473] introduced the point-by-point model by considering the emission from all individual fragments, specified by Z and A. This model was further developed e.g. by Lemaire et al [474], Tudora et al [475] and Vogt et al [476]. In [477–480], the spectral shape was parametrized by the Watt formula [471] or an empirical shape function that had been introduced by Mannhart [481] in order to better model the shape of the neutron energy spectra in the fragment frame. Kornilov [482] proposed a phenomenological approach for the parametrization of a model-independent shape of the prompt-neutron spectrum. This approach was later also used by Kodeli et al [483] and Maslov et al [484]. Recent refinements of the Los Alamos model were proposed in [224]. These are (i) separate contributions of the (average) light and the (average) heavy fragment to the prompt-neutron emission, (ii) the departure from statistical equilibrium at scission, (iii) the separate nuclear level densities for the fragments, and (iv) the center-of-mass anisotropy in the angular distribution.

The Watt model [471] and the Los Alamos model [330] are directly fitted to the measured prompt-neutron spectrum, while the point-by-point model [473] is based on the measured A-TKE distribution. Manea et al [485] proposed a scission-point model that provides the TKE(A) distribution, in order to allow calculations of prompt-neutron spectra with the point-by-point method, if only the mass distribution is known. Recently, several models have been developed that treat the statistical de-excitation of the fission fragments by prompt neutrons and prompt-gamma radiation in a more elaborate way, partially by the Hauser–Feshbach formalism. We mention here MCHF [216], CGMF [218], FIFRELIN [220], and FREYA [221, 222]. A special feature of these models is the most accurate reproduction of the prompt-gamma spectrum, in particular the peak structure at low gamma energies, mostly produced by the non-statistical gammas emitted at the end of the gamma de-excitation cascade based on the detailed tables of experimental spectroscopic information of the fission fragments.

5. Discussion and outlook

After this detailed report on the different activities in fission research, we will try to give an assessment of the status and the most important achievements that have promoted the understanding of nuclear fission during the last few years. This will lead to an outlook on the developments to be expected in the near future and on the challenges ahead to be tackled.

5.1. Status of microscopic theories

There is no doubt that microscopic theories are indispensable for a deeper understanding of the fission process. However, in spite of considerable progress and many important results, the
theoretical description of the fission process with dynamical microscopic models is still very difficult, because the most advanced models in nuclear physics that have been developed for stationary states in heavy nuclei [486], for example modern versions of the interacting shell model or effective field theories of quantum chromodynamics [487], are not readily applicable to the decay of a meta-stable state. Intense efforts are presently being made to develop suitable theoretical tools, see [81] and section 4.1. Another difficulty arises from the technical limitations. Still, the application of the most advanced models, which are based on classical stochastic or self-consistent quantum-mechanical methods, is heavily restricted by their high demand on computer resources. In this section, we list some of the most important conceptual and technical challenges which these theories confront.

5.1.1. Restrictions by limited computer resources.

Number of relevant degrees of freedom. For the microscopic quantum-mechanical approaches based on TDGCM and for classical stochastic models, the number of relevant degrees of freedom that are presently explicitly treated are insufficient for a realistic dynamical calculation of the fission process and for covering the full variety of fission observables. In these microscopic and stochastic models, the number of relevant degrees of freedom is presently limited to two—respectively four or five—in the most advanced approaches. The success of the random-walk approach of Randrup, Möller et al in a five-dimensional deformation space [18] in reproducing the mass distributions of a great number of fissioning systems seems to indicate that the number of relevant degrees of freedom is very important for obtaining realistic fission-fragment distributions. Their model allows for fully independent shapes of the two nascent fragments. This elaborate feature seems to be more important than the restrictions to a comparably simple handling of the dynamics, assuming over-damped motion and using Metropolis sampling, as long as fission-fragment mass distributions are concerned. However, the calculation of other fission quantities, like the charge polarization (N/Z degree of freedom), fission-fragment excitation energies or their angular momenta requires the inclusion of an extended set of relevant degrees of freedom. Recently, the charge polarization degree of freedom was also included [55]. Progress is expected to come gradually via the continuous development of computer technology.

In the TDDFT approach (e.g. [68]), the need to construct collective variables, potential energy surfaces and the corresponding mass tensor as a pre-requisite for the dynamical calculations is obviated. Only the starting point of the TDDFT calculation in terms of suitably chosen constraints on the potential-energy surface needs to be determined, as discussed in section 4.1. Thus, the above-mentioned restrictions are overcome. However, this advantage is restricted by a considerably increased computational cost which presently limits the applicability essentially to the calculation of the most probable fission quantities and a few very specific problems. Neglect or approximate treatment of effects beyond the mean field. Another class of difficulties arises from the effects beyond the mean field in microscopic quantum-mechanical models. The handling of many-body dynamics is very difficult, and, thus, much effort has been put into developing suitable approximate algorithms that do not demand too much computer resource (see e.g. [289]). Questions regarding how well the physics is still represented must be answered.

5.1.2. Problems in determining the potential-energy surface. For self-consistent microscopic approaches (see section 4.1) and for the stochastic models (see section 4.2), the calculated potential energy in the space defined by the relevant degrees of freedom is the basis for the dynamical calculation of full distributions of fission observables. There are several difficulties associated with the determination of this multi-dimensional potential-energy surface.

Usually, in stochastic models a shape parametrization is used, and the potential-energy is calculated with the macroscopic–microscopic approach. In this case, only a restricted class of shapes can be realized. This means that the calculated potential energy is the upper limit of the optimum potential that the nucleus can adopt. The deviations can be reduced by increasing the dimension of the deformation space. However, as said before, the tractable number of relevant degrees of freedom, in particular in a dynamical calculation, is restricted due to the limited computing resources, as seen in section 5.1.1.

When the potential energy is determined self-consistently with constraints on some degrees of freedom, this is a safe method for finding the optimum shape. However, the optimum shape is determined independently on the different grid points defined by the constraints. This can lead to discontinuities in the shape evolution from one grid point to the next, because one or several of the degrees of freedom that do not belong to the ones explicitly considered may take very different values. This means that the ‘real’ nuclear potential may have a ridge that is not visible in the calculation [77, 297]. Again here, the small number of dimensions aggravates the problem.

Another class of problems occurs in case a shape parametrization, generally used in stochastic models, an optimization with respect to the additional shape degrees of freedom is performed individually on each grid point that does not belong to the degrees of freedom explicitly considered in the dynamic calculation [377].

It is important to control the effect of such problems on the result of the dynamical calculation, if they cannot be avoided. The appearance of such problems can be detected by local unphysical fluctuations [297] in the calculated potential-energy landscape. It can be reduced by an optimum choice [306] and by increasing the number of relevant shape parameters.

As mentioned before, this problem does not exist when the TDDFT is used to compute the dynamical evolution of the nucleus from an initial point beyond the second fission barrier up to scission [68, 95], because the shape of the system develops freely in a self-consistent way without any constraint.
5.2. Aspects of statistical mechanics

Most of the models applied to nuclear fission consider aspects of statistical mechanics only by global properties of the degrees of freedom that are treated as an environment. This is, firstly, the level density of the fissioning system, and secondly, dissipation by the coupling between the relevant degrees of freedom and the environment. Phenomena that are connected with the energy transfer between subsystems of the environment, as described in section 4.3.6, are most often not considered.

At present, stochastic models are able to treat dissipation by global descriptions of one-body [488, 489] and two-body [313] mechanisms and to include the effects of pairing correlations and shell effects on the transport coefficients and the level density for determining the heat capacity of the environment, and for deriving the driving force of the fission dynamics [83].

Self-consistent quantum-mechanical models have only recently overcome the restriction to adiabaticity and methods have started to develop that enable the consideration of quasi-particle excitations on the fission path [15] and one-body dissipation by the fast shape changes at neck rupture, e.g. [47].

Energy exchange between the nascent fragments [12], or even the competition between quasi-particle excitations in the neutron and proton subsystems [490] of the fragments, were only considered in dedicated approaches. However, for an understanding of the division of excitation energy between the fragments or the odd–even effect in fission-fragment nuclide distributions, the explicit consideration of the two environments in the two nascent fragments and their interaction is indispensable.

Preliminary results on these rather complex features of statistical mechanics with simplifying assumptions have already been obtained. These concern the phenomenon of energy sorting [12] and the global features of the odd–even staggering in fission-fragment Z distributions [52].

The division of excitation energy between the fragments has recently attracted quite some attention. The energy dissipated separately in individual nascent fragments on the fission path was estimated by Mirea [16] and compared with the experimental data. The division of excitation energy between the fragments induced at neck rupture was studied in the sudden approximation [24]. An interesting attempt to study the energy partition between fragments with a microscopic self-consistent approach was performed in [22] by considering spatially localized quasi-particles in a frozen scission configuration. The dominant role of statistical mechanics, and particularly the assumption of statistical equilibrium in the division of heat between the nascent fragments before scission, which is made in [12, 228], is questioned [22] or criticized [68] by several authors. But the remarkable experimental result of [413] in which an increased initial excitation energy of the fissioning system is found in the heavy fragment, while the neutron multiplicity of the light fragment stays unchanged, has not yet been addressed by microscopic models. This is also true for the complex features of the odd–even effect in fission-fragment Z distributions, which is also successfully described in the framework of statistical mechanics in [52].

5.3. Systematics and regularities

In section 4.3 we presented the combinations of several empirical observations and powerful theoretical ideas as a basis of the GEF model. They go well beyond purely empirical descriptions, because they do not only reproduce experimental data with high accuracy, but, due to their theoretical basis and the relatively small number of adjustable parameters that describe all systems with identical values, they are also expected to provide reliable predictions for a large variety of fission quantities over a wide range of nuclei for which no experimental data exist. In particular, it was demonstrated in section 4.4 that the results of GEF agree very well with very different experimental data that have not been used to fix the parameters of the GEF model. The GEF model code [228] that exploits these ideas pursues the tradition of former inventive ideas like the macroscopic–microscopic model [298] and the concept of fission channels [244] and, partly, makes direct use of them.

The relationship between GEF and microscopic fission models may best be illustrated by recalling the role of the liquid-drop model in the development of nuclear mass models, although the dynamical fission process is much more complex than the static properties of a nucleus in its ground state. For a long time, purely microscopic models were not able to attain the accuracy of the liquid-drop model in reproducing macroscopic nuclear properties. Only very recently has the accuracy of fully microscopic and self-consistent models become comparable with the accuracy of macroscopic–microscopic models [365, 491]. While the powerful basic relations of the liquid-drop model follow directly from the theoretical assumption of a leptodermous system, the parameter values were determined by an adjustment to experimental masses and other nuclear properties. Only microscopic models were able to relate the values of these model parameters to the properties of the nuclear force [492]. Remembering this analogy clarifies that GEF is not intended to compete with microscopic models, although it is presently better suited as far as the use for applications is concerned. In contrast, both approaches may be considered to be complementary for extracting physics. In particular, the semi-empirical GEF model helps to uncover regularities that are not directly evident from the fission observables and to recognize the physics content of some systematic trends in the data.

5.4. Uncomprehended observations

Beyond the general inability of theory to explain many facets of the nuclear-fission process, there are some specific observations that seem to contradict the well established knowledge. In the following, we will present one of those cases that we believe to be among the most striking.

5.4.1. Dominance of ‘magic’ proton numbers in fission-fragment distributions. The success of the GEF model [228] in reproducing the fission-fragment mass distributions from the fission of actinides by a statistical approach, assuming the universal fragment shells superimposed on the macroscopic potential, is already a remarkable result. Even more striking
is the constancy of the mean number of protons in the heavy fragment of the contributions from the asymmetric fission channels standard 1 and standard 2 over all systems investigated until now [273]. In particular, for the standard 2 fission channel, this finding seems to contradict the results of Strutinsky-type calculations [342] within the macroscopic–microscopic approach performed by Wilkins, Steinberg and Chasman [248], who attributed this dominant asymmetric fission channel in the actinides to a shell at a large deformation in the neutron subsystem at $N = 88$. The deformation of about $\beta = 0.6$ that they found in their calculations is consistent with the neutron multiplicity observed in the heavy fragment. These calculations did not provide any evidence for a proton shell at $Z = 55$, which one might suspect to be responsible for the fixed mean number of protons found in the experiment. Other systematic Strutinsky-type calculations performed by Ragnarsson and Sheline [493] yielded similar results.

Until now, Randrup, Möller et al [32, 154] performed the most extended systematic calculations of the fission-fragment mass distributions for a large number of fissioning systems with the Brownian Metropolis shape-motion treatment. In a recent study, the mean number of protons in a heavy fragment was investigated in dedicated calculations [89] for a series of uranium isotopes ($^{234}$U, $^{236}$U, $^{238}$U and $^{240}$U), and the experimental results, available for $^{234}$U, $^{236}$U and $^{238}$U, were rather well reproduced. This model has the potential to shed some light on the prevalence of proton or neutron shells, because it is able to explicitly calculate the neutron shell plus pairing corrections and the proton shell plus pairing corrections. However, [89] does not give any detailed information on this question. Furthermore, it is difficult to establish a relation to the hypothetical role of the fragment shells assumed in the GEF model [228], because the fission channels standard 1 and standard 2 are not distinguished in [89].

One might imagine a number of reasons for the observation of a nearly constant number of protons in a heavy fragment of the asymmetric fission channels under the assumption of the decisive role of fragment shells. Some possible explanations could be (i) that the relation between the size of the shell-stabilized pre-fragment and the size of the final fragment is not so strict, for example by a variable division of the number of nucleons in the neck at scission, or (ii) that the Strutinsky-type calculations of [248, 493] are not realistic and miss a proton shell near $Z = 55$ at a large deformation. In this context, we would like to recall the observation of a mutual support of magicities in the surrounding of spherical doubly magic nuclei [494], because it violates the independence of shell effects in the neutron and proton subsystems. A similar effect in deformed nuclei may be expected. Maybe, the position of a shell in the proton or neutron number even moves as a function of the number of nucleons of the other kind. However, such an effect is not observed in the case of some local stabilizations by neutron shells at $N = 152$ (see e.g. [372]) and $N = 162$ (see e.g. [241]), which are stable in neutron number over several elements.

In any case, the almost constant number of protons in the heavy fragment, found in the contribution of the most important asymmetric fission channels in the actinides, is a very intriguing observation that asks for a deeper understanding. Interestingly enough, the position of the shell in the light fragment to which the double-humped mass distributions in the lead region are tentatively attributed shows the same feature, see section 4.3.5.

5.5. Modeling of fission dynamics

In section 4, the different approaches and their methods of modeling fission dynamics have been described separately. In this section, we would like to make a survey on the strengths and limitations of each of these methods for treating fission dynamics in a comparative consideration. We will discuss the time-dependent density-functional theory (TDDFT), the time-dependent generator-coordinate method with a Gaussian overlap approximation (TDGCM-GOA), and stochastic models. Finally, we will also give a short overview of the semi-empirical GEF model.

TDDFT provides the most elaborate modeling of the shape evolution of the fissioning system. It allows for all kinds of shapes without any constraint, and thus, all collective degrees of freedom (CDOF) are included. All shapes are allowed, and the nucleus chooses the path in the shape space dynamically. The forces acting on the nucleons are determined by the nucleon distributions and velocities, and the nuclear system naturally and smoothly evolves into separate fission fragments. It was found that the strong energy exchange between a large number of CDOF slows the rolling down towards scission and leads to a very long fission time [68]. One could say that the collective degrees of freedom are strongly coupled between each other and form a kind of special heat bath, not in equilibrium with intrinsic excitations. As discussed in section 4.1, one-body dissipation and some quasi-particle excitations are already included in TDDFT. The authors of [68] state that extensions of the present approach to two-body observables [20] (fission-fragment mass, charge, angular momenta and excitation-energy distribution widths) are rather straightforward to implement, and, eventually, stochasticity of the mean field could be introduced. However, these calculations are still very much hampered by the immense demand on computer resources, severely aggravated by the fact that the distributions of fission quantities are constructed by accumulating the results of many fission events (the Monte-Carlo method) [95].

In TDGCM-GOA, the dynamics is calculated on a potential-energy surface, presently spanned by two collective parameters (for example two multipole moments). The shapes and binding energies are calculated independently for the different points of the potential-energy grid. This makes it impractical to handle the energy transfer between the differently shaped degrees of freedom or the excitations of collective modes. Moreover, it is necessary to impose a scission configuration with a somewhat arbitrarily defined criterion. In addition, the presently available calculations assume adiabaticity.

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20 According to [68], these observables cannot be determined properly by considering only one-body interactions (the interaction of independent nucleons with the common potential). Also two-body interactions (interactions between two nucleons) must be included.
The inclusion of dissipation, leading to intrinsic excitations (single-particle or quasi-particle excitations) is just being developed.

An elaborate consideration on the prospects and the limitations of the different self-consistent approaches has been formulated by Bulgac [495].

Advanced stochastic models calculate the dynamics on a potential-energy surface, presently spanned by up to five dimensions. Like in the TDGCM-GOA method, the binding energies are calculated independently for the different points of the potential-energy grid. Due to the shape parametrization, only a few of the collective degrees of freedom are included. Therefore, it is again unpractical to handle the excitations of all the collective modes and the direct energy transfer between different CDOF. Intrinsic degrees of freedom are included via a representative heat bath with a certain temperature. The heat bath introduces stochastic fluctuations in the trajectory in the collective-coordinate space. It is also the most effective mean for the (indirect) transfer of energy from one CDOF to another. Note that this scenario does not agree with the direct coupling between the collective degrees of freedom found in TDDFT [68].

In the GEF model, the excitations of collective and intrinsic degrees of freedom are only included in an effective way by the temperature values of the relevant environment. Depending on the coupling strengths between the intrinsic and collective degrees of freedom, or between the collective degrees of freedom alone, this could be the temperature of the heat bath that represents the intrinsic degrees of freedom, like in the stochastic models, or it could be the average energy per collective degrees of freedom, which is suggested by TDDFT. GEF may act as a bridge between microscopic models and empirical information, by the values of the temperature parameters that describe the population of the normal oscillators or the widths of the distributions of the respective fission quantities, see equations (8) and (9). GEF is presently the only model that explicitly considers the heat transfer between the nascent fragments as a collective degree of freedom.

Obviously, the different models are presently either not yet applicable or conceptually unable to treat all aspects of fission dynamics. TDDFT is the only approach that includes all collective degrees of freedom. However, it is still under development in many aspects and generally very much hampered by the high demand on computer resources. TDGCM-GOA is unpractical in handling the excitations of all collective degrees of freedom and lacks the inclusion of all kinds of intrinsic excitations. Stochastic models handle the coupling between collective and intrinsic excitations in the most simple and practical way. However, the excitation of collective degrees of freedom is very restricted, and the assumption of thermal equilibrium between collective and intrinsic degrees of freedom is a strong and probably wrong assumption. The GEF model contributes to the idea that heat storage should be considered separately for the two nascent fragments. This emphasizes that aspects of statistical mechanics should be taken into account in realistic modeling of the fission process.

5.6. Theoretical needs

In this review, many restrictions of the different theoretical approaches to nuclear fission were quoted. In the following, we summarize the expected evolution of fission theory to satisfy the main identified needs.

5.6.1. Extending the relevant degrees of freedom. With the progress in computer technology, the corresponding restrictions will gradually become less severe. First of all, this will allow the number of relevant degrees of freedom to be extended in the stochastic and the microscopic quantum-mechanical models that explicitly use a potential-energy surface. For example, it will become customary for the shapes of the two nascent fragments to be allowed to evolve independently, and the charge polarization will routinely be considered. These developments are important for handling fission-fragment distributions in a more realistic way, fully specified in mass and atomic number [55].

Moreover, progress is needed to clarify the processes that are responsible for the generation of the angular momentum of fission fragments and the eventual generation of orbital angular momentum [496], as well as the evolution of the projection of the total angular momentum onto the symmetry axis of the fissioning nucleus [497]. Also, the excitation of other collective modes, which are already the subject of dedicated theoretical considerations, and some stochastic calculations may be studied by microscopic models.

There are strong arguments that the intrinsic and collective degrees of freedom of the environment form separate thermodynamical units with different temperatures [68, 246]. Furthermore, to describe the heat transport between the nascent fragments, it appears to be mandatory for the part of the environment which comprises the intrinsic degrees of freedom to be divided into two parts, belonging to each fragment. The properties of these partial environments should be treated separately, and their interaction should be considered.

5.6.2. Effects beyond the mean field. More powerful computing resources may also allow the application of more realistic treatments of the effects beyond the mean field than those that are manageable at present, see section 4.1.

5.6.3. Dissipation. While dissipation—the coupling between the relevant degrees of freedom and the environment—is an inherent part of stochastic approaches, microscopic quantum-mechanical models were originally restricted to adiabatic processes. Algorithms for considering dissipative processes are presently being developed, for example, by explicitly including the first quasi-particle excitations [15] or by representing the heat bath of stochastic models by a large number of identical quantum oscillators [498] (independent oscillator model [499]). There is great interest in developing more realistic and more complete representations of the environment that better reflect the complex nuclear properties.
5.6.4. Evolution from the mononuclear to the dinuclear regime. The gradual transition from the mononuclear to the dinuclear regime of two nascent fragments that are coupled by the neck manifests itself in several ways: the early predominance of fragment shells seems to be well established [242, 368], but also the localization of quasi-particles in the two fragments [424] and the increase of the congruence energy [500] need to be better understood. The increase of the pairing gap is also expected to evolve gradually during the fission process. The odd–even staggering of the fission barrier demonstrated in section 4.3.1, which is not correctly reproduced—e.g. [364], can be understood as a sign for the shape dependence of the pairing gap.

5.6.5. Neck rupture. Realistic modeling of the violent processes around scission is very demanding for the treatment of collective dynamics and the induced intrinsic and collective excitations. In principle, TDDFT masters this problem to some extent, but the calculation of the corresponding fission observables, like the odd–even effect in fission fragment Z distributions or the generation of fragment angular momentum, is not yet possible. Progress in the self-consistent modeling of quantum localization and other processes around neck rupture is expected to improve the understanding of the instabilities at neck rupture and their effect on different observables, like the odd–even effect in fission-fragment Z distributions and in the kinetic energies, or the angular momenta of the fission fragments.

5.6.6. Combination of different approaches. Progress in the modeling of nuclear fission may also evolve from exploiting the strengths of methods from different approaches. For example, elaborate stochastic models provide the technical tools for handling dissipation, which are directly considered in the Langevin equations by the friction tensor and the fluctuating term. Even further, the driving force $T dS/dq$ is essentially determined by the derivative of the entropy—a quantity that is primarily a property of the environment. It has been mentioned that the microscopic models do not treat fully quantum mechanically intrinsic and collective degrees of freedom and their coupling. But without considering in some way the environment that creates the driving force, friction and fluctuations, a realistic description of the dynamics is not possible. A solution could be to use quantum-mechanical considerations for estimating the mass tensor [62], the full friction tensor and the dependence of the entropy on the relevant degrees of freedom, including the excitation energy, and to perform the dynamical calculation with a stochastic approach. A step in this direction has already been made [79]. However, the Langevin equations are based on the assumption that all degrees of freedom of the environment form a heat bath, which means that they are in statistical equilibrium at every moment. This is probably not always a realistic assumption, and it might be necessary to take this into account.

The transformation of energy and the transport of heat between different subsystems during the fission process are genuine problems of statistical mechanics. It would constitute great progress if considerations of the statistical mechanics could be introduced into microscopic quantum-mechanical approaches in some manageable way.

The observation of gradual systematic variations of the fission quantities along a series of neighboring nuclei may be exploited to increase the accuracy of microscopic models that treat each fissioning system independently. Each system has its own technical uncertainties, which are inherent, for example, in the shell effects determined by the Strutinsky procedure or in the potential energy determined by a restricted deformation space, discussed in section 5.1.2.

One may also extend the validity range of semi-empirical approaches like the GEF model, which covers a considerable variety of fission quantities, if one succeeds in deriving the justification for some approximations of the GEF model as well as for the values of the model parameters on a microscopic basis.

5.7. Experimental needs

In the field of nuclear fission, which is a process that is still so far from being fully understood, it is not possible to give a list of the most important missing experimental information. One should be prepared for surprises and for new problems to emerge when new data come up. However, some general rules may be given. It is certainly beneficial to cover a range in the choice of the fissioning system in terms of the nuclear composition (Z and A), excitation energy and angular momentum that is as wide as possible. The angular momentum dependence can be investigated by forming the same fissioning nucleus with different reactions. Moreover, the coverage of the fission observables should be as complete as possible, and it should be measured with the best resolution and with as many coinciding quantities as possible.

In the following, we will illustrate the relevance of the rules mentioned above by recalling the progress brought about by some specific experimental information and the open questions that could be answered by new measurements.

5.7.1. Wide coverage and precise definition of initial excitation energy.

Distinguishing different fission chances. At energies that are above the threshold for multi-chance fission, the fission fragments are emitted from a wide range of excitation energy. Up to third-chance fission (the fission after the emission of two neutrons), step-like structures can be observed in the fission cross section. These carry some information on the different contributions. However at higher initial excitation energies, these data do not provide unambiguous information on the evolution of the fission-fragment mass distributions and the fission probabilities. Theoretical estimates have a large margin of uncertainty [330, 501]. See, in particular, figure 27 of [501], where the uncertainty of the relative weights of the different fission chances are demonstrated. Further development of suitable differential techniques [502] to distinguish the fission events from different fission chances would improve the
experimental knowledge on the washing out of shell effects in the fission probability and in the fission-fragment production with increasing excitation energy at fission. Valuable information on multi-chance fission can also be deduced from the relative number of neutrons emitted before and after fission, or more exactly before and after passing the fission barrier. However, measuring the number of neutrons emitted from the fully accelerated fragments, which is easily determined by a multi-source fit of the prompt-neutron angular distribution, is not significant, because neutrons emitted from the first minimum cannot be distinguished experimentally from those emitted between the barrier and scission. Energy dependence of odd–even effect in different observables. The experimental information available at present on the decrease of the odd–even effect in fission-fragment Z distributions with increasing initial excitation energy has been obtained with relatively broad excitation-energy distributions, e.g. by bremsstrahlung-induced fission [503], or by electromagnetic-induced fission at relativistic energies [274]. The first results on the energy dependence of the odd–even effect in fission-fragment N distributions have only been deduced very recently by comparing the new data from electromagnetic-induced fission at relativistic energies [116] with the results from thermal-neutron-induced fission, see section 3.3. The determination of the energy-dependent odd–even staggering in fission-fragment Z and N distributions, the total kinetic energies and other observables would certainly help us better understand the influence of pairing correlations on the fission process.

5.7.2. Extended systematic coverage of fissioning systems.

Shell structure in fragment distributions around $A = 200$. The observation of a double-humped mass distribution in the fission of $^{180}$Hg [252] drew attention to the appearance of structural effects in the fission of lighter fissioning nuclei ($A < 210$). In spite of the intense experimental effort, it has not yet been possible to establish a comprehensive overview on the mass distributions of these nuclei in low-energy fission. The most efficient method to provide a wide systematics of fission-fragment distribution for fission from energies in the vicinity of the fission barrier of neutron-deficient systems over a large mass range is the electromagnetic-induced fission of relativistic projectile fragments that is presently being used by the SOFIA experiment [192]. This approach will even provide nuclide distributions, fully separated in $A$ and $Z$, which allow a much better characterization of the fission process than the mass distributions with a limited resolution obtained from direct-kinematic experiments. New experimental results are urgently awaited.

Fragment distributions for long isotopic chains. The evolution of fragment mass distributions is of great interest for two reasons: firstly, it will help us better estimate the mass distributions from the fission of very neutron-rich nuclei on the astrophysical $r$-process path. This information is urgently needed so as to be able to simulate nuclide production in the $r$-process. Secondly, the position of the mean $Z$ in the heavy component of the asymmetric fission channels could be followed over a larger range. This would help us better understand the mechanism behind the surprisingly almost constant mean $Z$ values in the heavy components of the fission-fragment distributions for actinides, and the weak variation of the proton number of the light peak from fission in the lead region.

5.7.3. Correlations of as many observables as possible. The small amount of data on the variation of the mass-dependent prompt-neutron multiplicities as a function of initial excitation energy is presently the only rather direct experimental evidence for the energy-sorting process (see section 4.3.6). This illustrates the importance of multi-parameter experiments for discovering new features in the fission process. This concerns, for example, the identification of fission fragments in $A$ and $Z$ and the measurement of the kinetic energies, multiplicities and energies of prompt neutrons and prompt gammas. In general, such data will provide important constraints on the modeling of fission.

5.8. Possible scenario of the fission process

5.8.1. Proposed scenario. On the basis of a synthetic view of experimental information and of the different theoretical approaches and ideas, we propose a possible scenario of the nuclear-fission process. The following description is rather comprehensive in covering the whole fission process, but not complete in all details.

We consider a system that starts from a configuration in the first minimum with a certain excitation energy $E^*$ above the ground state. On the way to fission, the excited nucleus has to cross the fission barrier first, which has a height of at least 3 MeV, for the nuclei considered in this work. In this case, the passage of the barrier is governed by the following characteristics: Let us first consider the evolution of the intrinsic excitation energy $E^*$ (or the entropy $S$, which is strictly connected to $E^*$ in the case of statistical equilibrium among the intrinsic degrees of freedom). If the entropy is preserved, the system can only proceed to fission by tunneling through the fission barrier. Tunneling through the whole barrier is strongly hindered by a factor of roughly $10^{-15}$ or more [372]. Therefore, it is very probable that the system will convert part of its excitation energy into deformation energy in order to pass the barrier without tunneling—if the initial excitation energy allows it. The density of transition states above the saddle (or above the inner saddle in the case of a multi-humped barrier) is lower by a factor of about $\exp(-3/T) = 0.0025$ (using a typical value of $T = 0.5$ MeV for $A \approx 200$ [394]) or smaller, compared to the density of states in the first minimum. According to the transition-state model [504], there is a high probability ($> 0.9975$) that the single-particle configuration of the system will change before the system reaches the saddle. There is no preference in populating any state at the barrier with the suitable energy, angular momentum and parity. Therefore, the distribution of transition states accumulated over a large number of fission events corresponds to the statistical equilibrium of the system at the given energy [373]. This means that
there is no memory of the initial state in which the system was formed in the first minimum, or of any state before reaching the outer barrier, except excitation energy, angular momentum and parity. Some peculiarities should be mentioned: When the transition states at the given energy are resolved, the equilibrium distribution of these states eventually shrinks to a single state. When the initial excitation energy is lower than the fission-barrier height, the system can only pass the barrier by tunneling. This occurs most probably when the intrinsic excitation energy is zero both at the entrance point before and at the exit point beyond the barrier. In the case of a double-humped barrier, the passage of the second barrier essentially proceeds with the same characteristics. For nuclei with a double-humped (or eventually with a triple-humped) barrier, this means that it fully adapts in all its degrees of freedom to the phase space above the outer-most barrier.

Beyond the outer-most barrier, the system is driven to scission and, finally, to disintegration—in most cases into two fragments—by the dominant influence of the Coulomb force. On the way from the outer saddle to scission, the system experiences a very complex evolution. In addition to the intrinsic excitations, the system has full freedom in its shape evolution. (In a simplified way, the intrinsic excitations are often represented by a heat bath, while the shape evolution is often represented by the elongation and a number of normal modes like the octupole moment that determines the mass asymmetry and neck radius of the fissioning system, the quadrupole moments of the two nascent fragments, etc. The normal modes may be represented by harmonic oscillators with their minima at the static fission path, the characteristics of these harmonic oscillators evolve with elongation, and the different normal modes may be coupled to each other. For example, the mass asymmetry couples strongly with the quadrupole moments of the two nascent fragments.) In this dynamic process, the driving force along the fission path drives the system continuously out of equilibrium, and full statistical equilibrium cannot be attained any more. Several forces are acting on the system: the shape evolution is described by a kind of transport equation with appropriate coupling to the intrinsic degrees of freedom, including many-body interactions; consideration of quantum mechanics is mandatory. (In the simplified picture mentioned above, the motion towards increasing elongation is coupled to the other shape degrees of freedom. Moreover, the coupling of the shape degrees of freedom to the heat bath of intrinsic degrees of freedom induces entropy-driven forces and dissipation.)

A few quantum-mechanical features, which are connected with the residual interactions (that go beyond the independent-particle model or the mean-field approximation), are known to be subject to a considerable change during the fission process. In particular, this concerns the total pairing condensation energy and the total congruence energy. It is expected that both quantities change their magnitude continuously during the evolution from a mononucleus to a dinuclear system [424, 500]. It is also expected that other properties of the fissioning system, like the shell effects [242, 368] and the level density, will approach the superposition of the properties of the nascent fragments more and more with gradually decreasing coupling between them. The latter leads to the transport of thermal energy between the nascent fragments and—to the extent to which statistical equilibrium is realized—to the phenomenon of energy sorting [12].

After scission, the fragments attain the shapes of their respective ground-state, experience some transformation of energy from deformation energy and collective excitation energy into intrinsic excitation energy, eventually experience some Coulomb excitation in the field of the complementary fragment [439], and are accelerated to their final velocities, while conserving their total energies, angular momenta and parities.

After scission, the excitation energy and angular momentum difference with respect to the the ground states of the separate fragments are carried away in a statistical de-excitation process. On a longer time scale, the \( \beta \)-unstable fragments are subject to delayed processes that are, among others, responsible for the emission of anti-neutrinos, delayed neutrons and the decay heat of used fuel.

5.8.2. Confrontation with theoretical models. The theoretical approaches, described in section 4, do not fully cover the above-mentioned scenario and partly show more or less severe deviations. We will mention only a few of the most important ones, and we concentrate on the evolution of the system from the barrier to scission, which is the main subject of most theoretical models.

Among the microscopic self-consistent approaches, the TDGCM with GOA assumes adiabaticity during the evolution from saddle to scission, considers only a very limited number of collective modes, and cannot model the scission process. The proposed TDDFT approaches handle only some limited aspects of dissipation. Tanimura et al [95], based on TDHF-BCS, starts the dynamic calculation around mid-way between saddle and scission with an arbitrary distribution of initial conditions and treats the effects connected with pairing correlations in the BCS approximation, which includes the pairing effects essentially only on static nuclear properties. In its concept, TDHF-B [68] agrees with the proposed scenario to a great extent, although dissipation is only partly considered, and the effect of residual interactions beyond pairing and the heat transfer in the dinuclear regime are not considered—at least not explicitly. In addition, the predicted quantities are still very limited.

The stochastic models do not include quantum-mechanical effects in the dynamic evolution and only consider the motion in a limited number of collective coordinates by entropy-driven forces as well as dissipation in the interaction with the heat bath. Furthermore, only a limited number of normal collective modes is considered.

The semi-empirical GEF model [228] is guided by the proposed scenario, although the physics is not always explicitly formulated and often expressed by effective descriptions. The importance of statistical mechanics and the early localization of the nuclear wave functions in the two nascent fragments before scission are particularly emphasized. These aspects are not explicitly considered by the other models mentioned before. The model owes its wide coverage of fission quantities
6. Summary

The experimental and theoretical activities of the last few years that have most strongly promoted the understanding of nuclear fission have been covered in this review, along with prospects for future developments.

On the experimental side, the application of inverse kinematics has extended the experimental capabilities in several aspects. At present, this is the only way to identify all fission products in $Z$ and $A$. In an approach developed at GSI, Darmstadt, fragmentation products from a relativistic $^{238}\text{U}$ beam were fully identified in $Z$ and $A$ and brought to fission by electromagnetic excitation in a heavy target material. With this technique, the low-energy fission of a large number of nuclei with $A \leq 238$ that were not accessible before can now be investigated. The fission products can be identified in $Z$ and $A$ with an excellent resolution. This technique has already proven its potential by mapping the transition from symmetric fission to asymmetric fission around $^{226}\text{Th}$. Furthermore, the first results demonstrate that this technique also offers unique possibilities for systematic experiments on lighter neutron-deficient nuclei in an extended region around $^{180}\text{Hg}$ [171]. In another approach, developed at GANIL, Caen, the transfer reactions of a $^{238}\text{U}$ primary beam in a carbon target have given access to experiments on fission for a number of heavier nuclei with well-defined excitation energy and the separation of all fission products in $Z$ and $A$. At CERN-ISOLDE, the progress in LASER ionization has made it possible to study beta-delayed fission with fully identified ISOL beams and to discover the asymmetric fission of $^{180}\text{Hg}$ as well as the structural effects in the mass distributions of other neighboring nuclei.

However, these measurements are still essentially restricted to fission-fragment yields and total kinetic energies. Extensions that include the measurements of prompt neutrons and prompt gammas in coincidence with the fragments would be very important for a better understanding of the energetics of the fission process and of the generation of the fragment angular momenta.

On the theoretical side, much effort has been invested in developing the microscopic quantum-mechanical self-consistent descriptions of the fission process. In spite of the difficulties caused by the high demand on computer power, the lack of suitable tools for handling non-equilibrium processes and the difficulties of introducing phenomena related to statistical mechanics to a quantum-mechanical description, progress is being made in promoting a qualitative understanding of the fundamental aspects of nuclear fission. The dynamical TDDFT approach, which avoids any constraint on the collective variables, is among the most interesting of recent developments.

The stochastic description of the fission process by the numerical solution of the Langevin equations, after being successfully applied for many years to study high-energy fission, has recently also been applied to low-energy fission, where shell effects and pairing correlations play an important role. The strength of this approach is in the inherent treatment of statistical mechanics, but the drawback is the classical character of the Langevin equations and the assumption of a uniform heat bath. Systematic dynamical calculations of the fission quantities and their variation as a function of the nuclear composition and the excitation energy are possible. Unfortunately, the necessity for Monte-Carlo sampling entails a limitation of the number of relevant degrees of freedom that are explicitly considered and, thus, a restriction in the coverage of fission quantities, or a strongly simplified treatment of the dynamics. A gradual extension is expected in line with the progress of computer technology.

Another possibility for modeling fission consists of a combination of powerful theoretical ideas and empirical knowledge. A rather successful example is the recently developed GEF model, which is based on a global view on experimental findings and the application of a few general theorems, rules and ideas. It covers almost all fission observables and is able to reproduce measured data with high accuracy while having remarkable predictive power by establishing and exploiting unexpected systematics and hidden regularities in the fission observables. This model has revealed features that are not covered by current microscopic and self-consistent models, in particular several manifestations of statistical mechanics. A highlight is the discovery of energy sorting.

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