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Review

The most important theoretical developments leading to the current understanding of heavy-element stability

With some personal recollections from the past 55 years (1965–2020)

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Abstract We discuss the sequence of developments that over the past 90 years led to current insights on heavy-element stability. The semi-empirical mass model, and its extension to deformed shapes, developed in the period 1936–1950 allowed the interpretation of nuclear fission. Around 1950 the spherical single-particle model was developed, soon after with extension to deformed nuclei. Speculations about a shell-stabilized region of spherical heavy elements near $Z = 126$ were made. In the 1960ies Strutinsky combined the single-particle and macroscopic liquid-drop models into a unified picture, the shell-correction, or macroscopic-microscopic method. Now it was also realized that although $Z = 126$ was present, an often stronger spherical gap in calculated proton single-particle level diagrams, $Z = 114$, was also present, but its significance had previously been overlooked. A large number of studies of the stability of nuclei in the "shell-stabilized" region surrounding $Z = 114$ and $N = 184$ followed. Initially the assumption was that elements just beyond the actinides, would be too unstable to be observed. The 1970ies saw considerable work in refining the initial single-particle and macroscopic models. This set the stage for global studies, which took off in the 1980ies and have continued until today. The more accurate nuclear-structure models allowed calculations of masses, decay-chain properties and branching between different decay modes to useful accuracy and predictive quality. A completely unexpected result was that the calculations showed the existence of an area of relatively stable deformed nuclei in the presumed "sea of instability" between the actinides and the next postulated spherical magic numbers.

1 Introduction

The limits to the number of different elements that can exist is set by (in)stability with respect to nuclear fission. In nuclear fission nuclei decay by splitting into two fragments of roughly the same size. Stability of nuclei with respect to this decay mode decreases with increasing proton number. Current impressions are that the number of elements is limited to about 120. At this time (2022) elements up to proton number $Z = 118$ have been identified and named. However, for a nuclear theorist it is also of interest to understand isotopic stability, that is the stability versus neutron number of each element. Here much is still unknown.

After the discovery of the neutron by Chadwick [1] in 1932 many experiments were performed, in which elements, in particular heavy elements, were bombarded with neutrons and the decay chains following neutron capture were observed. The expected decays of the nuclei formed were alpha-decay, beta-decay and electron capture, possibly a few in succession. The anticipation, or bias, was that the decay products would be elements and isotopes with proton and neutron numbers relatively close to the target nucleus. The possible decay modes and energy releases were usually calculated by use of the semi-empirical mass model in the form proposed by Bethe and Bacher [2]. However a confusing number of "decay periods" were observed. This confusion reigned until November 1938–February 1939 when two monumental developments occurred: (1) The definite identification of barium in the decay products following neutron bombardment of uranium [3] which led (2) Meitner and Frisch to propose that after neutron capture the nucleus, instead of emitting a particle, deforms as a liquid drop and divides into two fragments of roughly equal size [4]. These fragments would then repel each other with very high kinetic energies. A few weeks after this idea emerged Frisch measured these high

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fragment kinetic energies [5], which definitely confirmed the deformed liquid-drop interpretation.

2 1936–1949: macroscopic liquid-drop model era

To show that a very simple model can be of enormous importance[1] we present some details of the original "liquid-drop" model. The first version of the liquid-drop model was actually not (yet) a liquid-drop model but a spherical semi-empirical mass model. In this first global macroscopic nuclear-mass model of immense practical utility the nuclear ground-state mass is given by

$$E_{\rm mac}(Z,N,{\rm shape}) = M_{\rm H}Z + M_{\rm n}N - B(N,Z)$$

where $M_{\rm H}$ is the mass of the hydrogen atom and $M_{\rm n}$ that of the neutron. The nuclear binding energy $B(N,Z)$ is in the model by Bethe [2] and Weizäcker [6] written as

$$\begin{aligned} B(N,Z) = & \\ & +a_{\rm v}A && \text{(Volume energy)} \\ & -a_{\rm s}A^{2/3} && \text{(Surface energy)} \\ & -a_{\rm C}\frac{Z^2}{A^{1/3}} && \text{(Coulomb energy)} \\ & -a_I\frac{(N-Z)^2}{A} && \text{(Symmetry energy)} \\ & -\delta(A) && \text{(Pairing energy)} \end{aligned}$$

Meitner and Frisch proposed that fission could be pictured as a charged "liquid drop" undergoing a sequence of shape changes and subsequent division into two fragments of somewhat similar size [4]. During this shape change the Coulomb- and surface-energy terms are not given by the expressions above, which are appropriate only for a spherical shape, and therefore need to be modified to account for deformation effects. The other terms are independent of shape. Bohr and Wheeler in their 1939 seminal paper [7] described the deformation dependence in terms of Taylor expansions. For a full discussion we refer to their paper. To show that spherical (macroscopic) nuclei become unstable to fission at high proton number we only need the lowest-order terms in their description.

Let the nuclear surface be described by

$$r(\theta,\phi) = R_0\left[1+\alpha_2 P_2(\cos\theta)\right]$$

Then the surface energy to lowest-order Taylor expansion is:

$$E_{\rm s} = E_{\rm s}^0\left(1+\frac{2}{5}{\alpha_2}^2\right)$$

and the Coulomb energy to lowest-order Taylor expansion

$$E_{\rm C} = E_{\rm C}^0\left(1-\frac{1}{5}{\alpha_2}^2\right)$$

The energy at deformation α_2 relative to spherical shape is

$$E_{\rm def}(\alpha_2) = E_{\rm C}(\alpha_2) + E_{\rm s}(\alpha_2) - (E_{\rm C}^0 + E_{\rm s}^0)$$

If $E_{\rm def}$ is negative then the spherical shape has no barrier with respect to shape change and is consequently unstable with respect to fission. The condition is

$$E_{\rm def}(\alpha_2) = \frac{2}{5}{\alpha_2}^2E_{\rm s}^0 - \frac{1}{5}{\alpha_2}^2E_{\rm C}^0 < 0$$

It is customary to define a fissility parameter x and write this condition as

$$1 < \frac{E_{\rm C}^0}{2E_{\rm s}^0} = x$$

With parameters used at the time [6] the surface energy for a sphere is given by

$$E_{\rm s}^0 = 17.80A^{2/3}$$

and the Coulomb energy for a sphere by

$$E_{\rm C}^0 = 0.7103\frac{Z^2}{A^{1/3}}$$

Thus the fissility parameter x is:

$$x = \frac{Z^2}{50.13A}$$

In Table 1 we give values of the fissility parameter x for a few values of Z and A. It is clear stability with respect to fission limits the number of elements that can exist. The root cause is actually a simple consequence of the long range of the Coulomb force and the short range of the nuclear force.

This was put on a firm quantitative basis in the paper by Bohr and Wheeler [7]. Based on their theoretical considerations they also argued that ^{239}Pu should be more fissionable than ^{235}U.[2] This was soon confirmed by Seaborg and collaborators [8–10]. In their large-ranging investigation Bohr and Wheeler carried their Taylor expansions to higher order and provided barrier estimates for nuclei throughout the periodic system. Somewhat later Frankel and Metropolis in a paper [11] carried out modern numerical integrations of the

[1] Stanislav Ulam has observed: It is still an unending source of surprise for me to see how a few scribbles on a blackboard or on a sheet of paper could change the course of human affairs (http://yquotes.com/quotes/stanislaw-ulam/).

[2] We use here the ingrained terminology in the field, namely that in this context "^{235}U fission" actually refers to fission after a thermal neutron has been captured, that is fission of ^{236}U. In other contexts "fission of ^{235}U" may refer to fission of precisely ^{235}U. So it is extremely important to be aware of context to interpret a discussion correctly.

Table 1 Fissility parameter x for select Z and A. When $x > 1$ the spherical shape looses stability with respect to (spheroidal) deformation and there is no stability with respect to fission. However, for a nucleus to be observable in experiment the spontaneous-fission half-life needs to be in the order of nanoseconds or longer, which implies barriers must be higher than about 4 MeV. The liquid-drop model picture is modified by microscopic corrections

Z	A	x
50	124	0.402
82	208	0.645
92	138	0.709
100	252	0.792
114	298	0.870
125	328	0.950
130	335	1.006

Coulomb and surface-energy terms in the liquid-drop model on the ENIAC computer. In particular they showed how saddle-point shapes and barrier heights depend on the fissility parameter x in the range from $x = 1$ to about $x = 0.65$, namely spherical for the heaviest systems and more elongated for lower x. The results of these pioneering studies have held up well in later investigations. During this era several new elements were discovered. Also experimental data on level structure in nuclei started to accumulate.

3 1950–1959: single-particle model era

The Bethe and Bacher (BB) paper [2], introduced the idea that non-macroscopic effects could lead to differences between measured masses and the semi-empirical mass-model values and that at least some of these differences could be understood in terms of shell structure seen in calculated single-particle level schemes; BB used a spherical oscillator potential to find such gaps. In particular, large gaps in the calculated level spectra would lead to particularly strongly bound nuclei. They stated that they could find significant differences between the semi-empirical mass-model value and the measured mass of ^{16}O. For heavier nuclei there were not sufficiently accurate data at the time to form any conclusions. In their words: "It seems in fact that there is ample evidence for a particular stability of ^{16}O, and thus for the individual-particle approximation." So shell-effects is not a new concept although their magnitude could not be calculated until 30 years later. At the time the general consensus was that a spin-orbit force would be very weak so there was no need to include such terms in single-particle models.

However, when sufficiently accurate experimental data on masses became available, showing increased stability for proton and neutron numbers 2, 8, 20, 28, 50, and 82, and neutron number 126, the higher "magic numbers" did not correspond to gaps in the oscillator-potential level structure. Moreover, spectroscopic data showed a large splitting between spin-orbit partners $j = l + 1/2$ and $j = l - 1/2$ with a sign so that the latter one has the higher energy. Such a model gave the observed magic numbers as well as in many cases the observed odd-particle spins as discussed in Refs. [12–15]. Some deviations between predicted spins and experimental data did exist. Mayer mentioned in particular $^{23}_{11}\mathrm{Na}_{12}$ and $^{55}_{25}\mathrm{Mn}_{30}$ [14] and tries to explain it within the spherical single-particle model.

It turned out that to obtain a general single-particle model of nuclei the assumption of a spherical-shape potential well had to be generalized to include deformed single-particle potential wells. Rainwater based his proposal to consider deformations on the observed large quadrupole moments of nuclei [16] and some simple theoretical calculations. Soon after, Bohr and Mottelson developed in extensive detail a unified model of nuclear properties and incorporated both microscopic and macroscopic ("liquid-drop model") aspects [17,18]. Because of insufficient computer power at the time it was impossible to solve for levels as exactly as would be desired. However, Nilsson took a decisive step forward with his thesis work "Binding States of Individual Nucleons in Strongly Deformed Nuclei" [19]. But computer power was still so limited he had to ignore couplings between shells of different main quantum number N in the oscillator potential he was using.[3] In this paper he introduced (in appendix A) another representation in which matrix elements of the quadrupole operator and other terms in the Hamiltonian between N and $N + 2$ were either zero or so small that they could be ignored. Subsequently this representation was referred to as the *"stretched"* representation [20]. Later a large-scale comparison to experimental data, such as odd-A spins, magnetic moments and ground-state deformations, were carried out by Mottelson and Nilsson [21].

A problem was how to calculate the ground-state deformations. Often several methods are explored in the same paper. To study if the deformed single-particle model agrees with experimental spins some authors used measured quadrupole moments to deduce a deformation and compare calculated levels at that deformation to experiment. Others looked at experimental spins and determined which calculated shape would yield levels in agreement with observations. In Ref. [21] calculated energies of occupied single-particle levels were added up, the obtained values were plotted versus deformation and the minimum energy and deformation were deter-

[3] Sven-Gösta Nilsson (SGN) told PM that he calculated each matrix element by help of a mechanical calculator and Clebsch-Gordan tables, wrote down the calculated matrix elements on paper and delivered his hand-written matrices to the computer center which then cranked out the eigenvalues.

mined. This in practice worked quite well so it was subsequently used as late as 1967 [22] and 1968 [23]. However, it became increasingly evident that this method had serious drawbacks, some of which were actually discussed in Nilsson's thesis in 1955 [19]. It was only with the introduction of the Strutinsky shell-correction method [24,25] that a reliable method for calculating the potential energy as a function of shape almost overnight completely changed the field. The importance of this method cannot be overstated, it is still in extensive use today more than 50 years later.

Between 1940 and 1959 many new elements beyond uranium were artificially produced and their fission properties determined, such as spontaneous-fission half-lives. Swiatecki discussed that observations showed that spontaneous-fission half-lives varied irregularly with Z and A, which could not be understood in terms of the smoothly varying barrier potential energies obtained in the liquid-drop model. He proposed that "shell structure" at the ground state was the reason for this deviation [26]. He calculated the energy effect of shell structure as the difference between measured masses and the masses given by the liquid-drop model. With this approach he could explain in impressive detail the irregular behavior of the measured spontaneous-fission half-lives. [4]

Experimentally it had been observed that odd nuclei had enhanced spontaneous-fission half-lives compared with even nuclei, which was illustrated in the discussion in [26]. The mechanism behind this enhancement was discussed in terms of a "specialization energy", an enhancement to the fission barrier that depends on the ground-state spin of the fissioning system, with increasing enhancement with increasing ground-state spin [27,28]. Because of this enhancement the first isotope of a new heavy element that is discovered is usually odd.

The rapidly decreasing spontaneous-fission half-lives with increasing proton number seen in the experimental data (for example Fig. 1 in [26]) might at first glance indicate that few additional elements beyond proton number $Z = 100$ could be produced. However SharffGoldhaber pointed out [29]: "Relatively long-lived isotopes may well be found among the far-transuranic nuclei because of magic-number stability. There may be, for instance another region of relative stability at the doubly magic nucleus ${}_{126}X^{310}$ (the closing of the next neutron j-shell)". Because of the casual way this possibility is discussed PM can only conclude this was not a sensational new insight but that the possibility had been circulating in the community "since the beginning". Later $Z = 114$ was promoted as the most likely next proton "magic number" candidate. This development is one topic of the next section.

[4] PM asked Swiatecki "How was this idea of shell effects in deformed nuclei received by the community?" Swiatecki responded "Glenn Seaborg gave me a job." And indeed Swiatecki was subsequently pursuing research in Berkeley for the next 54 years 1955–2009.

4 1960–1969: shell-correction method and the dawn of the potential-energy "surfaces" era

One would perhaps have expected that rapid exploitation and investigations of deformed single-particle models would follow the developments in the 1950ies but the computer technology did still not exist to solve existing models to desired accuracy. For example in the caption to the proton single-particle level diagram in Fig. 5 in Ref. [21] it is stated: "In drawing the figure, the states of the $N = 6$ shell have been consistently plotted at an energy 2.4 MeV lower than corresponding to these parameters. This corresponds to using a μ values of approximately 0.62 for the $N = 6$ states in this diagram". So, apparently, computer resources at this time were so limited that the authors did an estimate rather than recalculate.[5]. But discussion of spherical super-heavy elements continued, initially still focusing on $Z = 126$ as the next spherical magic proton number beyond $Z = 82$.

However, in the middle of this decade $Z = 114$ emerged as the candidate for the next spherical magic proton number beyond $Z = 82$. Myers and Swiatecki state in Ref. [30] (submitted Sept. 1965) "In our mass formula we have included, for purposes of illustration, magic numbers at $Z = 126$ and $N = 184$" but continue later "The actual values of the magic numbers might be different; for example, we have recently learned (Meldner and Röper, private comm. 1965) that $Z = 114$, $N = 184$ is a possible candidate for a doubly magic nucleus (see also p. 269, [31])". The figure on page 269 in [31] is actually a reproduction of a deformed level ("Nilsson") diagram from 1959 in [21], Fig. 5, p. 52. In that level diagram numbers are printed at the large gaps at $Z = 82$ and $Z = 126$. However there is also a large gap at $Z = 114$ which is left unmarked. Much later in the proceedings (published 1967, no submission dates given) from the 1966 Lysekil symposium, a similar level diagram is presented in Fig. 3 of Ref. [22]. This level diagram is based on further developments of the modified oscillator model and the better computational possibilities now available. There numbers are given in the gaps at $Z = 82$ and $Z = 126$ but

[5] In Lund, location of one of the major universities in Sweden, the only computer between 1956 and 1970 was a computer called SMIL consisting of about 4000 vacuum tubes and memory limited to 40 Kilobit (https://kulturportallund.se/smil-lund-forsta-dator-var-sveriges-andra/) When it became operational it was claimed it would solve all calculational problems in Sweden for ever! PM ran programs on this computer as part of his undergraduate classes.

none at $Z = 114$, although the figure shows it is calculated to be about twice as large as $Z = 126$.[6]

In Ref. [32] (submitted July 22, 1966) calculated spherical level diagrams in a Woods-Saxon model are presented, with the $Z = 114$ and $N = 184$ gaps clearly visible and marked. However, based on the above discussion it seems the published record shows that it is Meldner and Röper who first realized that $Z = 114$ was a plausible candidate for the next magic number.

At the time of the new ideas about the next magic proton number beyond $Z = 82$ a development of monumental importance occurred. It had long been noticed that nuclear mass calculations based on some version of the macroscopic liquid drop model, sometimes referred to as the semi-empirical mass model, showed increasingly larger deviations from measurements with decreasing distance from magic numbers. These deviations had long been referred to as "shell corrections" [2] but usually only in the context of the nuclear ground state. In Ref. [30] postulated expressions with adjustable parameters were used to represent these shell corrections and a global mass table was calculated. Calculations of this type were sometimes referred to as macroscopic-microscopic models. We mentioned in the previous section that Swiatecki observed [26] that "shell corrections" to the ground-state minimum of the macroscopic liquid-drop fission barrier also had a large effect on spontaneous-fission half-lives.

However, these early ideas did not permit actual *calculations* of "shell corrections"; experimental data had to be used in some fashion to account for them. A practical, predictive way of calculating microscopic shell effects is due to Strutinsky in the form of his shell-correction method. Thus "microscopic" in the macroscopic-microscopic method could now be calculated not just for the ground-state shape but for any value of Z and N, and for any nuclear shape starting from the calculated energies of single-particle levels in a deformed potential [24,25]. In particular the potential energy could be calculated for selected sequences of shapes leading from the ground state to separated fission fragments. Strutinsky also adapted his ideas to the calculation of pairing-correction energies. Application of this method immediately suggested an explanation for the recently observed fission-isomeric states [33], namely that these represented local shape-isomeric minima of nuclei with ellipsoidal shapes such that the ratio of the lengths of the major and minor axes was approximately 2:1.

Many groups had over the previous decade developed single-particle models to study low-lying energy levels in nuclei. These were now immediately widely used to calculate microscopic shell-and-pairing corrections and applied to the calculation of ground-state masses, fission barriers and stability with respect to various decay modes with much focus on the postulated super-heavy region of stability. The first comprehensive such study is that of Refs. [20,34,35]. The possible existence of super-heavy elements is mainly limited by stability with respect to fission. An extensive review of the many early studies of SHE fission and other decay properties are in Refs. [36,37]. In general most investigations found a region of relatively stable nuclides in the vicinity of $Z = 114$ and $N = 184$.

5 1970–1980: model refinement era

The utility of the Strutinsky method motivated in the 1970ies work on many refinements to the constituent single-particle and macroscopic models. The Nilsson modified oscillator potential was and is still being used but a basic difficulty is that the diffuseness of the nuclear surface is simulated by the so-called l^2 term. This feature makes it difficult to select the related diffuseness and spin-orbit parameters, κ and μ in regions of unknown nuclei. Therefore Wood–Saxon-type single-particle models were often preferred and more often used; increased computer power also helped. At the Los Alamos National Laboratory the folded-Yukawa single-particle model was developed following the ideas of Ray Nix; its original form is completely specified in Ref. [38] with some additional tweaks and current model constants in Ref. [39]. It was immediately used to study super-heavy element stability in Ref. [40]. In presenting the evolution of the nuclear potential energy from a nuclear ground state towards separated fragments in terms of a one-dimensional series of shapes it was selected to give the results as functions of the distance between the centers-of-mass of the emerging fragments. To model fission half-lives a model for the related inertia is also needed. It is well-known that the nuclear vibrational inertia is much larger than given by a model based on irrotational flow. But it was realized that in the limit of separated fragments the inertia is the reduced mass of the fragments. This important observation had not previously been implemented in models of the nuclear inertia.

Therefore a semi-empirical model for the inertia with a shape-dependence such that it is much larger than the irrotational inertia for ground-state shapes but evolves towards the reduced mass for separated fragments was proposed [40], with one parameter adjusted to ^{240}Pu half-lives. This turned out to be a very realistic proposal of great practical utility and the inertia model was used in many subsequent investigations; some examples are in Refs. [39,42–45].

In the first calculations [38,40] based on the folded-Yukawa single-particle model the choice of single-particle parameters, such as the diffuseness a and spin-orbit strength

[6] PM recalls that during a discussion with SGN some years later SGN looked somewhat sadly at the level diagram in [22] and said: "why did I not put 114 there."

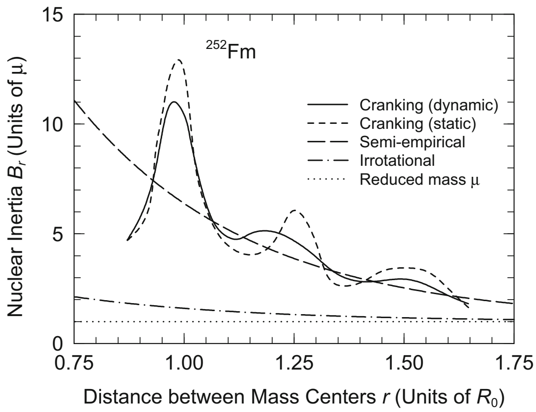

Fig. 1 Nuclear inertias in macroscopic and microscopic models. It is seen that the semi-empirical inertia follows closely the overall behavior of the cranking-model inertia. Also, towards the limit of separated fragments they approach the reduced mass, a necessary, known boundary condition. More details are in Refs. [40,41]

λ was based on a fit to levels in spherical nuclei in the Pb region made in Refs. [46,47]. About a year later SGN became concerned about the limited agreement between calculated and experimental low-lying single-particle energy levels in the deformed rare-earth and actinide regions.

Therefore new values were determined, one set for the rare-earth region and another set for the actinide region, with the diffuseness being the same in both regions [48].[7] Later, so that parameter values were available across the nuclear chart, the spin-orbit strength was postulated to depend linearly on A and the diffuseness to be constant [49] and this dependence was based on the values determined in Ref. [48] in the rare-earth and actinide regions. These single-particle parameters were always used subsequently by PM and usually by others performing calculations with the folded-Yukawa potential.

PM subsequently wondered if the new single-particle parameters had any significant effect on, for example, global mass calculations. Some years into the new century computers had become sufficiently powerful that it became possible, with fairly limited manpower, to investigate this. In Ref. [39] two global mass calculations are carried out. They are totally equivalent, with the one difference that in one the original choice [38] of single-particle parameters is used, in the other the new global set from [48,49]. With the new set of single-particle model spin-orbit and diffuseness strengths the model error decreased by 14.4%, see Fig. 36 in Ref. [39]. That a better reproduction of experimental level spectra simultaneously yields significantly better agreement with experimental nuclear masses is excellent support for the consistency of the Strutinsky shell-correction method. Furthermore we notice in the above-mentioned Fig. 36 that in the lower part with the original spin-orbit parameters that the mass-model error in the vicinity of ^{208}Pb is almost zero. That is probably because the parameters were tightly optimized to levels in ^{208}Pb in 1960 in Ref. [47], again illustrating that the more realistic the model level spectra are the better the calculated masses become.

The macroscopic model is also an important part of the macroscopic-microscopic model. Most calculations up to around 1970 had been content to use some minor perturbations of the original "semi-empirical mass formula" [2] with extensions to deformation. In the 1970ies there were two developments that took the original formulation in two somewhat different directions:

1. Myers and Swiatecki developed a systematic treatment of the macroscopic model to higher order in $A^{-1/3}$ and neutron excess $I = (N-Z)/(N+Z)$ in a series of papers [46,50,51], leading to the so-called droplet model.
2. In another direction Krappe and Nix and collaborators observed that the sharp-surface energy term in the original liquid-drop model would yield unphysically large surface energies for some configurations, for example fissioning nuclei with strongly necked-in shapes, or colliding heavy ions at close range. The first discussion of these ideas is in Ref. [52]. Further developments leading to the final version are in Ref. [53]. This model is designated FRLDM (finite-range liquid-drop model) which may refer either to the macroscopic model only or to the combination of the macroscopic model with folded-Yukawa single-particle shell and pairing corrections.

There has since the beginning of theoretical nuclear physics been a strong desire to develop theoretical models for nuclear masses since they define Q values of reactions and decays. The first such model, of enormous practical utility, was the semi-empirical mass model [2]. More realistic global (that is including nuclei from the lightest regions up to the super-heavy region and extending from the proton to the neutron drip line) models can be based on microscopic corrections calculated by use of shell corrections from realistic single-particle models with deformation taken into account. But they were slow in coming. The first such mass model is the one by Seeger and Howard. Details are in Ref. [54] and the table in [55]. It is the only contribution to the 1975 mass tabulations [56] where the microscopic contributions are calculated from levels obtained in a deformed potential well. None of the other contributions are based on a general nuclear-structure model that also provides other nuclear-structure quantities, in particular levels and deformations. Therefore, in such models, it is impossible to interpret deviations between calculated values and measurements in a way

[7] PM, at this time a graduate student, spent all of 1973 at Los Alamos and SGN half of his sabbatical there, the first part of 1973. They spent about 3 months of their time on determining new spin-orbit strengths and a new diffuseness parameter.

to yield useful information about paths forward to a more inclusive nuclear theory.

Fission studies were in this decade severely restricted by limited computer power. It was well known that many actinides divide asymmetrically preferably into one larger spherical fragment near ^{132}Sn and the remaining nucleons into a smaller deformed fragment. Thus, to study this process one should ideally calculate and study the potential-energy surface as a function of at least five independent shape variables, the obvious ones are: overall elongation (evolution in the "fission direction") two fragment deformations (ellipsoidal is a good first approximation), mass division between the two fragments and neck diameter. This would lead to calculations for hundreds of thousands or millions of different shapes, totally impossible for another couple of decades. It was hypothesized that the observed mass asymmetry in fission could begin to be established already at the second saddle (although Hill and Wheeler had earlier discarded this hypothesis in Ref. [57]). In Ref. [58] this idea was investigated, and it was found for the first time, that the outer saddle points were lowered by asymmetric shape-degrees of freedom for the selected actinide nuclei (^{236}U and ^{252}Fm) by up to 2 MeV. Due to the limited access to computer power the calculations only investigated 20 different shapes.[8] Pauli and collaborators a year later obtained similar results (but for ^{240}Pu based on a Woods-Saxon single-particle model) [59]. However, a later calculation [60] based on a calculation of 175 different shapes between ground-state and scission shapes for ^{236}U showed that the potential energy seemed to favor symmetric shapes at elongations beyond the saddle point. It was discussed in the paper that it might be an artifact of the limitation to 175 preselected deformation points and that a larger deformation space would show that asymmetry persisted from the saddle to scission. It would be another quarter century before increased computer power showed that this was indeed the case, see below.

However, the enthusiasm for super-heavy-element research started to wane as the decade progressed due to a complete lack of experimental evidence, as reviewed in 1979 in Ref. [61]. After the Ronneby Nobel Symposium on super-heavy elements [62] in 1974 even Sven–Gösta Nilsson told his then still large Lund group that "now we have to do something else" and for the brief remainder of his life[9] the group shifted focus to high-spin physics. PM participated in this change of direction, but spent two post-doc years (1975–1977) in Los Alamos. There he worked on heavy-ion reactions with emphasis on issues of importance for SHE production, and fission features. After the passing of SGN PM was invited for a three-month stay in Livermore followed by a five-month stay in Los Alamos through March 1980.

In Livermore PM together with W. Howard focused on calculations of fission barriers from the line of β-stability to the neutron drip line to provide input to r-process studies in particular where element synthesis might terminate, and if the r-process would be able to provide a pathway to the super-heavy region. After the Strutinsky method had been introduced this had quickly become a major focus area, some of the first such studies are in Refs. [63–67]. These studies all obtained a region of low fission barriers in the r-process path in a location with proton number larger than about 92 and neutron number larger than about 160 and seemed to exclude that super-heavy nuclei could be generated through decay from the r-process. However, Meldner had suggested that super-heavy elements could be formed in the neutron flux from timewise optimally spaced nuclear explosions [68–70]. The studies by PM and Howard in the summer of 1979 were based on the Lund modified oscillator single-particle model which had now been extended to mass asymmetric and axially asymmetric nuclear shapes. The host was Heiner Meldner[10] who took active part in many of the discussions of the results, but the calculations were carried out in collaboration with Michael Howard and published the next year in Ref. [71]. In the main features they agreed with earlier studies, namely that a region of low fission barriers might hinder super-heavy element formation in the r-process. In contrast to earlier results it provided an extensive and detailed theoretical nuclear-structure data base suitable for more detailed r-process studies.

In Los Alamos Ray Nix had taken notice of the Seeger and Howard calculation of a global mass "table" in [55]. In contrast to the other "tables" in the 1975 mass prediction [56], the calculation by Ref. [55] provided ground-state shapes, both quadrupole (ϵ_2) and hexadecapole (ϵ_4) moments and single-particle level diagrams as functions of shape. Ray Nix had invited PM for 5 months starting in October 1979, to work on the specific project of calculating nuclear masses based on the folded-Yukawa single-particle model and the finite-range macroscopic model of Ref. [53]. In the discussions of

[8] Although computer power was limited PM's funding (in Lund) to pay for computer hours used was four times his annual salary (which at the time was on par with the salary of an engineer at a private company! This was a deliberate government policy to encourage post-graduate studies.)

[9] SGN passed away April 24, 1979

[10] One of PM's most memorable experiences is being invited home for dinner to Heiner. We drove to the Livermore airport in his Ford Panterra, stepped into his plane (after a thorough pre-flight inspection), and lifted off. We flew straight above SFO airport (he had a transponder in his plane). PM asked, "can you really do that?" Answer: "It is a free country". We landed on an abandoned WWII air strip just half a mile from his house on a cliff overlooking Half-Moon Bay. After dinner and a sleepover we went for a walk along the cliffs in the early morning, had breakfast, flew back to Livermore and drove back to the laboratory. The normal daily commute of Heiner!

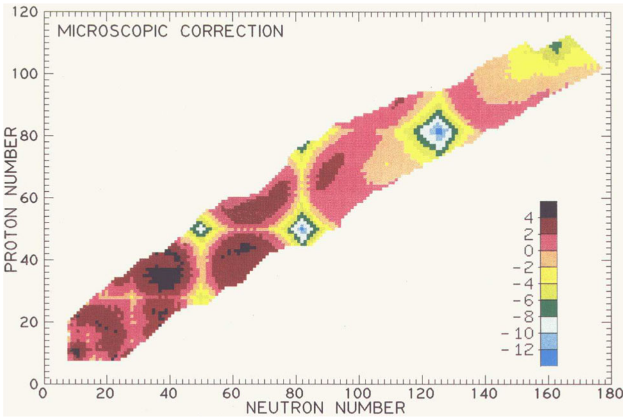

Fig. 2 Calculated microscopic contributions to the nuclear ground-state mass excess calculated in a folded-Yukawa model in Ref. [73]. The enhanced stability at doubly magic nuclei is obvious, unexpected was the deformed region of enhanced stability in the vicinity of $Z = 108$ and $N = 162$. The color plot of this calculation was first published in Ref. [74]

the scope of the calculation PM said that just as in the fission calculation with Howard [71] are we going to go the neutron drip line and how far above $Z = 114$ and $N = 184$ should we go? Ray Nix remarked

1. No, we do not want to blindly let the computer churn out numbers before we have done more tests of the reliability of the model so let us just extend along constant A four nucleons beyond the last known nuclei towards proton rich and neutron rich, and
2. let us not include the SHE region. There has been so much hype but no positive results for 15 years, so we do not want to give the impression that this is just another super-heavy element prediction; it is a mass table.

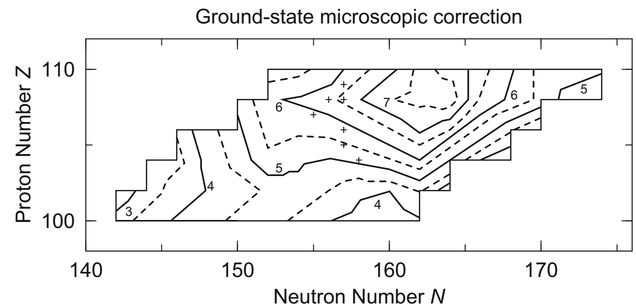

Fig. 4 Calculated microscopic contributions to the nuclear ground-state mass excess calculated in a Woods-Saxon potential in Ref. [76]. They are very similar to our results in Fig. 2

In hindsight this was being too conservative, see below about the super-heavy region. Because of this strategy the community had to wait for another 15 years for a published mass table

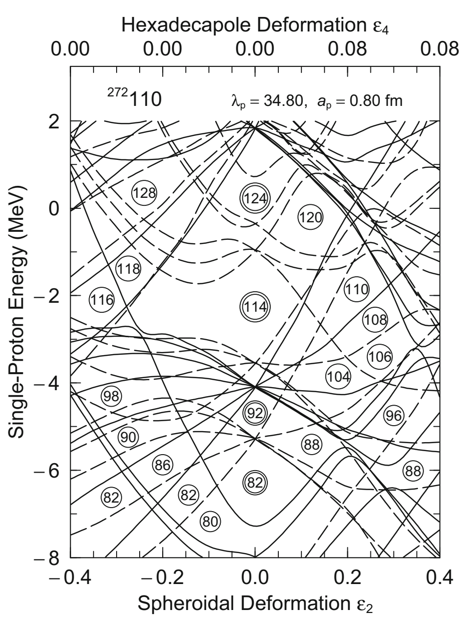

Fig. 3 Proton single-particle level diagram for nuclides in the vicinity of ^{272}Ds. The large gaps for deformed shapes for proton numbers in the range 104–110 stand out. The figure was first published in Ref. [75]

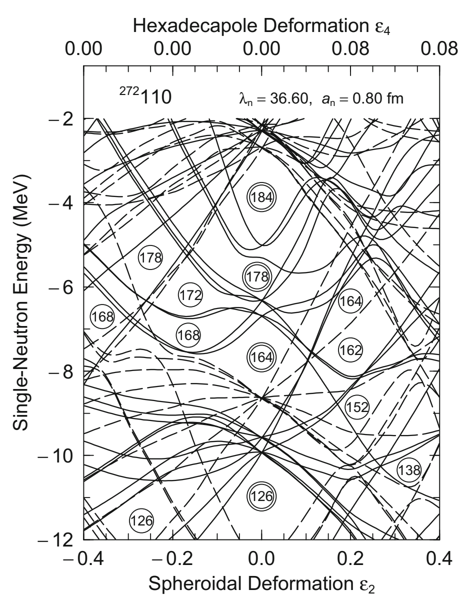

Fig. 5 Neutron single-particle level diagram for nuclides in the vicinity of ^{272}Ds. The large gaps for deformed shapes for neutron numbers 162 and 164 stand out. The figure was first published in Ref. [75]

to the drip lines [72].[11] Seeger and Howard [54,55] in the light nuclear region did not consider nuclei with $Z < 20$ and $N < 20$ because the macroscopic model as well as the mean-field single-particle model were not expected to be applicable to such light systems. In their calculations Möller and Nix assumed the same but implemented a variable lower cutoff and carried their calculations to ^{12}C "just in case". It was found that the model could be used to model nuclei down to $Z = 8$ and $N = 8$ so the lightest nucleus included in the mass table is ^{16}O. The calculations were completed in February 1980, see the next section for further discussion.

6 1980–2000: systematic global studies era

The mass calculation discussed above was completed in February 1980, and is described in detail in Ref. [49] in the Nuclear Physics **A361** No. 1 issue which was dedicated to the memory of Sven Gösta Nilsson. It has been designated as FRLDM1981. We just note here that the shape parameters varied in the search for the ground-state minimum were limited to quadrupole (ϵ_2) and hexadecapole (ϵ_4) deformations, due to the available computer power at the time. Interestingly there is very little mentioning of super-heavy-element stability in the **A361** issue. The mass table corresponding to the model in Ref. [49] was simultaneously published in Atomic Data and Nuclear Data Tables [73].

Many mass "tables" have been presented over the years. But an accepted view of "what is a scientific model" is that it should reproduce new data (in this case masses measured in the future) and ideally other types of data in addition to what it was initially focused on, in this case not just masses, and furthermore lead to unexpected new insights. The FRLDM1981 conforms to these 3 criteria.

In Ref. [49] it is mentioned that after the calculation was completed newly measured masses of isotopes of rubidium, cesium and francium were published. For six rubidium masses the rms error was 1.1 MeV, for seven cesium isotopes the rms deviation was 0.58 MeV and for ten francium isotopes the error was 0.48 MeV.

As regards new unexpected insights it is mentioned in the paper that nuclei in the vicinity of ^{222}Ra were calculated to be unstable with respect to octupole (ϵ_3) deformations. It had long been known that nuclei in this region exhibit low-lying negative parity states, so this led to more specific studies in Refs. [82–84]. More than 20 years later computer power had become sufficient to investigate instability with respect to octupole deformations globally in a full 4-dimensional space ($\epsilon_2, \epsilon_3, \epsilon_4, \epsilon_6$) see Refs. [85,86]. There was close overlap between regions where calculated instability with respect to octupole asymmetry occurred and the regions where low-lying negative parity states were observed experimentally. In addition, in the regions of calculated octupole instability, the consideration of this degree of freedom greatly improved the agreement between calculated and measured masses (Fig. 3 in Ref. [85]).

Perhaps the greatest "unexpected new insight" gained from the FRLDM1981 calculation occurred soon after its publication. At a lunch on the LBL cafeteria patio overlooking San Francisco Bay, Peter Armbruster pulled out a copy of the FRDM1981 mass table [73] and pointed to the result for $Z = 108$ and $A = 270$ which listed a ground-state microscopic correction of -6.04 MeV and similar values for nearby nuclei and asked PM "Do you think this has something to do with our discoveries of the new elements?". We show the 1981 calculated microscopic corrections as a color nuclear-chart type plot in Fig. 2, which plot was originally published in Ref. [74].[12] The region of large negative microscopic corrections centered at $Z = 108$ and $N = 162$ clearly stands out. We present in Fig. 3 a calculated "Nilsson-type" folded-Yukawa proton level diagram, appropriate for this region of nuclei, where the large deformed gaps for 104–110 are obvious. Similar results for shell corrections in this region were subsequently obtained in the Woods-Saxon potential, we show in Fig. 4 results from Ref. [76], redrawn by us and with text slightly modified to be consistent with current terminology. The calculated "Nilsson-type" folded-Yukawa neutron single-particle diagram for nuclei in this region is in Fig. 5.

It turns out, when we look back, that results showing large negative shell corrections in this region had been published earlier but their significance overlooked (just as was the case with the large gap at $Z = 114$, discussed above). For example in Ref. [20] Fig. 16 shows a calculated microscopic correction of about -5.0 MeV for $Z = 108$ and $N = 162$. which indicates stability with respect to fission might be sufficient to allow observations. In Ref. [48] table 2 shows a shell correction at $Z = 108$ and $N = 162$ of -8.05 MeV[13]. However, at the time nobody paid attention to these results because the focus was 1) to compare to known experimental data for

[11] The subsequent calculation was finalized in September 1992 and the mass table sent to Sigurd Hofmann at that time. The paper was submitted in the summer of 1993! PM thinks there was some debate at ADNDT whether they would devote a whole issue to what was submitted, because it was half a year before it was sent out for refereeing.

[12] It was very difficult to produce color plots of research results at this time. This figure is an exact (photo)copy of the printout in 1983 on an inkjet printer in development at the time by prof. Hellmuth Hertz at Lund University.

[13] One should note that "microscopic correction" is the sum of the shell correction at the ground state, zero-point energy and the difference between the macroscopic energy at the ground state and at spherical shape, so the microscopic correction for deformed ground states for nuclei in this region is higher by 2 MeV or so compared to the shell correction.

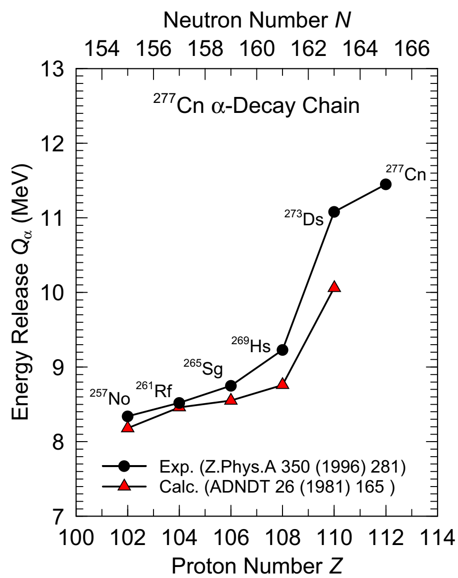

Fig. 6 Calculated Q_α values in the decay chain from ^{277}Cn determined from the mass excesses tabulated in Ref. [73] compared to experiment [77,78]. The kink in the experimental results verifies the calculated results (made 15 years earlier) pointing to a deformed region of nuclei with previously unanticipated large negative microscopic corrections

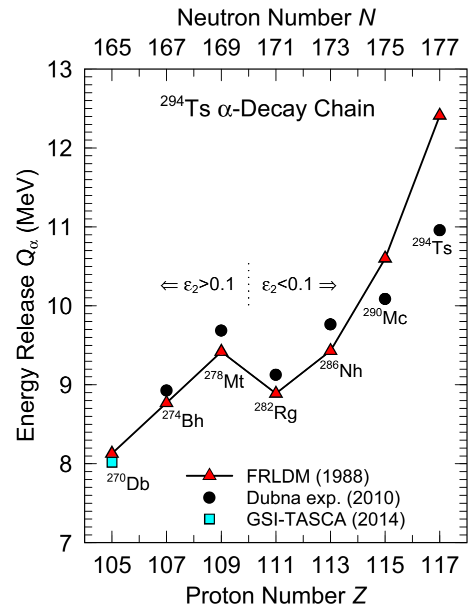

Fig. 7 Calculated Q_α values in the decay chain from ^{294}Ts from the mass model in Ref. [79] compared to experiments in Ref. [80], with the data point at $Z = 105$ from Ref. [81]

actinides with the aim to benchmark models and 2) in some studies to provide predictions about the postulated region of relatively stable spherical super-heavy nuclei. The observations by Peter Armbruster and the GSI results on $Z = 107$ [87] and soon after on $Z = 109$ [88] and $Z = 108$ [89] had the immediate consequence that George Leander (previously part of Nilsson's Lund group, but had some years earlier accepted a position at Oak Ridge) traveled to Los Alamos to run the codes of the FRDM1981 mass model to now actually calculate additional heavy "sea-of-instability" and spherical super-heavy masses so that results for a contiguous region from the actinides through the presumed "sea of instability" and including the spherical super-heavy region were consistently obtained in the finite-range folded-Yukawa model of Ref. [49,73]. Some results are in Ref. [90] with a full mass table of more than 4000 nuclei up to element $Z = 122$ in Ref. [79]. We compare the calculations in [73,79] of Q_α energies in decay chains from new elements discovered later. In Ref. [73] there are calculated mass excesses available to compare theoretical Q_α to the ones observed in the decay of ^{277}Cn [77], discovered 15 years after the prediction was made. Although calculated data for the first point in the decay chain is not provided in this calculation the remaining theoretical values are in good agreement. It is well known that "kinks" in Q_α energy curves in α-decay chains correspond to passing through regions of increased stability (negative shell corrections), usually magic numbers, but in this case a region of deformed nuclei with large gaps in the single-particle level spectra. The experiment is the first definite confirmation of the previously predicted deformed region of relatively stable nuclei.

In Fig. 7 we compare theoretical Q_α energies based on the mass table in [79] to the decay chain from the new element ^{294}Ts discovered 22 years after the theoretical predictions [80]. Again with encouraging agreement between observations and theory, keeping in mind that the observed decays occur in odd or odd-odd nuclei where the decays may not always be ground-state to ground-state, whereas the calculations refer to ground-state to ground-state transitions.

Similar, large negative shell corrections centered around $Z = 108$ and $N = 162$ were subsequently published based on a Woods-Saxon single-particle potential in Ref. [76]. Calculated fission barriers for a limited number of nuclei in this region, also based on the Woods-Saxon model, appeared in Ref. [91]. For nuclei in the heavy region with proton number somewhat above 106 the barrier height can also be simply estimated to be about the absolute value of the ground-state microscopic correction, that is around 5 to 7 MeV near $Z = 108$, $N = 162$, see discussion in Ref. [44]. Therefore overviews microscopic corrections such as those presented in figs. 2 and 4 are routinely used to identify nuclei that may be sufficiently stable to allow experimental observation. An early example is Fig. 16 in [20] mentioned earlier.

The "finite-range" macroscopic model [49,53] represents an effort to generalize a "standard" macroscopic liquid-drop model such as the one in Ref. [30] which has its origins in the Bethe-Weizäcker semi-empirical mass model and its generalization to deformed nuclei [7]. A development in a different direction was the so called “droplet model”. In that model the standard liquid-drop model is taken to higher order versus proton-neutron asymmetry $I = (N - Z)/(N + Z)$ and $A^{-1/3}$ [46,50,51,92]. In the early 1980ies Bill Myers suggested to PM that it would be interesting to incorporate the higher-order droplet-model effects *and* the finite-range effects of the nuclear force as consistently as possible in a single macroscopic model. Such an effort started, in collaboration with Swiatecki and Treiner when PM spent the summers of 1981 and 1982 as well as a sabbatical 1983–1984 at LBL. First results from this effort are in Ref. [93] in which a global mass calculation was performed with a resulting rms deviation of 0.676 MeV. It should be noted that the effects of ϵ_3 and ϵ_6 deformations were treated very approximately. This model is designated “finite-range droplet model” (FRDM) which expression may refer to the macroscopic model only or the combined macroscopic-microscopic model, depending on context.

A consistent mass calculation based on the FRDM and folded-Yukawa single-particle shell-and-pairing corrections was presented in Ref. [72]. This mass calculation is designated FRDM1992 because 1992 is the year when the calculation was completed and the mass tables made available on the LANL T-2 web site. Later it was observed that because the deformation dependence of some FRDM terms are treated in perturbation theory around spherical shapes, it should not be used in fission-barrier calculations. In the 1992 mass tabulations the ground-state shapes had been calculated in the FRLDM already in 1987 by PM under contract with LLNL. The computer resources in 1992 were still meager so the ground-state deformations were not recalculated in the FRDM model. The FRDM1992 mass table is therefore based on shapes calculated in the FRLDM. The same is the case for the FRDM2012 mass table [39] where the deformations are calculated in the FRLDM. It has been checked that the differences between the two models for the calculated ground-state shapes are negligible except that for the lightest nuclei the potential-energy surfaces are very flat in the FRDM with respect to all shape multipoles; therefore ground-state shapes would be difficult to determine in these cases. In the FRLDM the surfaces also become increasingly flat, but at a lower pace, with respect to higher multipoles towards lower A so the number of higher multipoles that can be investigated goes down as A decreases [96]. An intuitive way of looking at this is that the “wavelength” of the higher-multipole surface fluctuations should be comfortably larger than a nucleon size, a point also emphasized by Wilets in his book “Nuclear Fission” [97].

For the heavy-element region there is no “game-changing” improvement in mass-model accuracies since the FRDM1992 was submitted for publication. However, the FRDM2012 mass model shows significant improvements in some specific, localized regions outside the superheavy element region as is discussed in the publication [39].

Although measurements of Q_α values have been an important method to identify new heavy elements and isotopes it is fission that sets the limit to where the periodic system of nuclei terminates. To provide theoretical guidance it is therefore important to estimate spontaneous fission half-lives. In current approaches to calculate fission half-lives the results depend sensitively on the height of the barrier, the width of the barrier, the mass parameter associated with the shape evolution during barrier penetration and more. Early fission half-life calculations determined saddle points and minima in calculated potential-energy surfaces. Those were then used to construct one-dimensional barriers, an example is Ref. [42]. The mass parameter used there was the one proposed in Ref. [40]. Another approach, considered more sophisticated, is to calculate spontaneous-fission half-lives along various fission paths in a multi-dimensional deformation space with use cranking-model mass parameters as is done in Ref. [99].

Then the “minimum action” result provides the half-life. There are hundreds of different fission half-life calculations, but no dominating approach has emerged. One difficulty is that there is little guidance from experiments on what is the path through deformation space, what is the inertia at various points on the path, and what is the thickness of the barrier. Should one perhaps model the barrier penetration by considering an ensemble of one-dimensional barriers? After all fission terminates in an ensemble of different fragment divisions. The hundreds of different calculations of spontaneous-fission half-lives that have been published over the years therefore do not offer significantly more guidance on stability with respect to fission than a simple observation that in

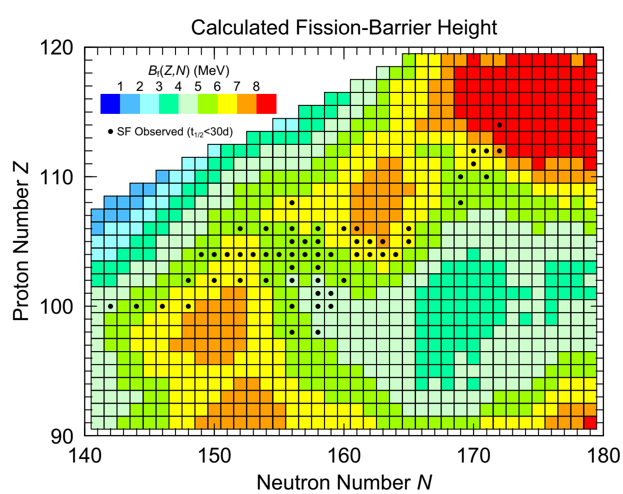

Fig. 8 Calculated barrier heights from Ref. [94]. We have marked with black dots those nuclides for which spontaneous fission with a half-life under about 30 days has been observed. The figure was first published in [95]

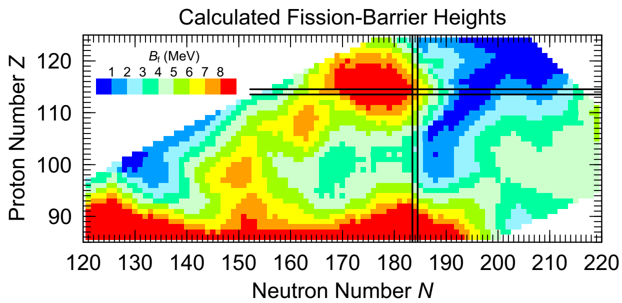

Fig. 9 Calculated fission-barrier heights in a folded-Yukawa model from Ref. [94]

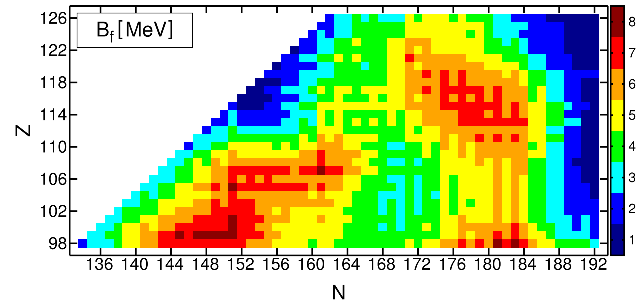

Fig. 10 Calculated fission-barrier heights in a Woods-Saxon model from Ref. [98]

the heaviest region where an outer barrier is largely absent the barrier height needs to be higher than about 5 MeV for the nucleus to be observable. The situation for understanding ground-state masses is much more favorable, because apart from masses, experiments can provide shape multipole moments and level structure at the ground state, which has been highly valuable for the development of theoretical models. Many of the quantities mentioned above that enter spontaneous-fission half-life calculations cannot be individually measured.

7 2000–2020: advanced fission modeling era

Because of vastly increased computing power it became possible to calculate fission potential-energy surfaces as functions of 5 shape variables, which number must be considered the minimum necessary, namely the ellipsoidal deformation of the two emerging fragments, overall elongation, mass division between the two fragments, and neck radius. Such studies, which provide potential-energy surfaces based on several million different shapes, resulted in vastly improved understanding of the fission potential energy as a function of shape. For example for some actinides there is in the potential-energy surface an obvious, persistent valley from an asymmetric saddle point to fragment separation [100–102]. Such a valley was not seen for ^{236}U in the early calculations in Ref. [60], but when the necessary five shape-degrees of freedom are taken into account the expected valley is present as was speculated in [60]. However, it is also possible for nuclei to fission asymmetrically without the presence of a persistent asymmetric valley from saddle to scission as is discussed in Refs. [103–105]. The more detailed calculations of potential energy surfaces led to advances in fission yield models [106,107], description of isotopic composition of fission fragments [108,109] and correlations between fragment total kinetic energies and neutron multiplicities [110] within a single model framework.

Although it has been stated [111] that the method of constrained Hartree-Fock potential-energy calculations makes it unnecessary to consider several independent (that is to constrain more than say two) shape variables in fission, this is not correct as discussed in [102], in more detail in [112] and from the HFB community perspective in Refs. [113,114]. Therefore the calculations discussed above based on energies calculated for up to ten million shapes yield insights not yet demonstrated in constrained HFB results.

For heavy nuclei in the superheavy region the outer peak in the barrier is absent or very low. It is important to realize that the most important point on the barrier is the ground state. If the ground state is lowered by say one MeV the barrier along its entire length becomes one MeV higher and the barrier also becomes somewhat wider. If the inner peak in the barrier is raised by one MeV the barrier height would be one MeV higher just as in the first case but only in a limited range of deformations, and there is no effect on the barrier width. Therefore the effect of lowering the ground-state energy by 1 MeV is roughly six orders of magnitude on the spontaneous-fission half-life, whereas raising the inner peak by the same amount will only affect the half-life by one or two orders of magnitude. It was already pointed out in Ref. [44] that for the heaviest nuclei the ground-state microscopic correction is the most important factor determining the stability with respect to fission, because for the heaviest nuclei its absolute magnitude is roughly equal to the barrier height.

In Fig. 8 we show calculated [94] barriers for a limited region heavy nuclei where we have marked with black dots those nuclides for which a spontaneous-fission half-life of less than about 30 days has been observed. Only a very few lie outside the range 5–7 MeV. Many nuclides in the plot are unknown, but we can fairly confidently conclude that if the barrier is below 5 MeV or so the spontaneous-fission half-life will be too short to permit observation. Further discussions are in Ref. [94]. There is no "go to" model of calculating spontaneous-fission half-lives (in contrast to ground-state properties). Some further discussion of spontaneous-fission half-life issues are in Ref. [41].

8 Prospects

During the professional career of PM 13 new elements have been discovered. For PM this has been an amazing journey because during the first 15 years of PM's career his advisor and PM were highly active in the field, but with null observations of superheavy elements as discussed above, which was very disappointing to a young researcher entering the field.[14] However, during this time computer power continually increased so that models could be vastly improved. When element 107 was discovered at the GSI the field was therefore ready for a synergetic collaboration between theorists and experimentalists. How many new elements will be discovered in the future? Another 13? PM does not think so. We show in Fig. 9 calculated barriers [94] from the actinides and extending beyond $Z = 120$ and $N = 184$. First, there is no hope beyond $N = 184$ except perhaps one or two nucleons beyond. Second, below $N = 184$ towards higher Z the barriers seem to offer some hope to go to a few additional elements. But here alpha decay will soon kill the prospects. So at best there seems to be no possibility of more than one or two more new elements. Very similar results are obtained in a Woods-Saxon barrier calculation [98] (but limited to a smaller region of nuclei), see those results in Fig. 10.

However, we should always remind ourselves that our models are based on simple "effective" forces. Several of the terms in the macroscopic models are simple Taylor expansions in neutron excess and not in our current implementations based on a fundamental model of, for example, symmetry energy behavior. Therefore, although they have performed "beyond expectation", there is always room for surprises. Valuable would be to in experiments further approach $N = 184$ to understand the strength of this postulated magic neutron number and to establish if the predicted and calculated "doubly-magic" spherical superheavy island really exists.

In concluding this exposé I like to express to my mentors, collaborators, and community, my immense gratitude for the more than half a century of amazing adventures in nuclear stability that I have been given the opportunity to participate in.

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Data Availibility Statement This manuscript has no associated data or the data will not be deposited. [Authors' comment: There are no associated data available.]

[14] When PM asked SGN in 1967 to be accepted for a Ph.D. program in his group, SGN looked very serious and said, "There is no future in theoretical nuclear physics". To which PM responded: "I just want to learn more physics, then I will search for a suitable job in industry".

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