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MY JOURNEY TO THE SUPERHEAVY ISLAND WITH SZYMANSKI, NILSSON, NIX, AND SWIATECKI FROM LYSEKIL TO THE PRESENT

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We discuss the great theoretical insights on nuclear stability that emerged in the late 1960ies, especially the realization that observable nuclei might include a superheavy island 20+ protons beyond uranium with 92 protons. However, we now realize that the early models were not sufficiently quantitative to yield definite conclusions. Further groundwork for such models were laid in the 1970ies. Around 1980 more quantitative, global and universal models appeared and together with experimental advances the mapping of the superheavy island, or as it turns out now, continent started in earnest. We review these early developments and conclude with some examples of our current insights. In particular, 1) some theoretical models do have predictive capabilities for nuclear properties such as ground-state masses, shapes, and half-lives 10 or more neutrons and protons away from previously known regions and 2) the superheavy island may be a continent connected to the actinides by a narrow land bridge; the continent itself may extend from about proton number $Z = 110$ to $Z = 120$ or slightly beyond.

1. 1960–1970: Emergence of a Superheavy-Element Vision

In this session in memory of Zdzislaw Szymansky let me start by recalling that I started my graduate studies in Lund with Sven-Gösta Nilsson in 1967, just at the time when Zdzislaw started his long, deep, and productive collaboration with the Lund group. Let me also mention that my first 5 papers are with Zdzislaw[1,2,3,4,5], although "my" is too strong, I was a very junior contributor at this time.

Three developments of enormous importance occurred just as I started my graduate studies. First, for the first time we had access to a computer which made serious (by the standard of the day) computing possible. We could for instance solve for the eigenvalues of a single-particle potential with matrix dimensions up to 80 or so. The first paper[1] exploits this resource to calculate hexadecapole components of the ground-state shapes of actinide and rare-earth nuclei. It also illustrates the mindset imparted on me by Zdzislaw and my other three mentors (in the title): *Data is king.* Both calculations of ground-state hexadecapole moments and their experimental determination were quite new, and we immediately embarked on a comparison between model results and measured data, discovering quite satisfactory agreement.

In this paper[1] the old method of summing single-particle levels to calculate nuclear potential energies as functions of shape was still used but this was soon to change, I never used it again, because of the other two monumental developments at this time. The year before, at the 1966 Lysekil conference, the second important development I alluded to above was the introduction to the world of the Strutinsky shell-correction method[6], the third the realization that new magic numbers beyond $Z = 82$, $N = 126$ could enhance stability of very heavy elements sufficiently so that they could be observed.

Let me step back for a moment so that we can remind ourselves that much, or most, fundamental insight into low-energy nuclear-structure physics have come from simple models and their extensive comparisons to data, provided they qualify as models. Principles of science tell us that a model needs some basic properties to qualify as a “model” rather than being a trivial parameterization of data: 1) it should also be applicable to data not used in the determination of its parameters, 2) it should be applicable to new types of data, and 3) it should be generalizable in new directions.

The very simple Bethe-Weizsäcker mass model with its 5 parameters was of enormous importance in the 1930ies in unravelling the mysteries of element transmutation. When fission was discovered, the model was immediately generalized to describe the potential energies of deformed nuclei and offered an unprecedented interpretation of the newly observed fission phenomenon[7]. When more extensive comparisons of nuclear masses to the model results became available, magic numbers were discovered. These in turn led to the formulation of the spherical single-particle model. Deviations between calculated and observed nuclear ground-state spins, plus a few other observations, inspired Nilsson to develop the *deformed* single-particle model[8]. This model explained a vast amount of spectroscopic data.

Swiatecki, in 1955, realized that microscopic effects could have a significant effect on the potential energy, not just near magic numbers for spherical nuclei but also in the ground-states of deformed nuclei. Again the insight came from looking at deviations between a simple model, the liquid-drop model of fission, and data. Swiatecki realized that the rapid variation of fission half-lives, especially near $N = 152$ could not be described by the transmission through a barrier as given by the liquid drop model. He also observed that the ground-state masses as given by the liquid-drop mass model were not as bound as the observed masses[9]. He therefore lowered the liquid-drop model fission-barrier ground-state minimum by the difference between the observed and theoretical masses, which led to an increase of the barrier height by variable amounts, compared to the macroscopic liquid-drop barrier. This was the seed of the shell correction method. However, it could only be applied when the experimental masses were known, and only at the ground state.

Strutinsky’s contribution, of monumental practical importance, and intellectual ingenuity, was that he provided a method by which the shell correction could be *calculated* for *any* shape and *any* single-particle potential. This was first presented to a larger audience at the Lysekil conference[6]. The possibility of relatively stable superheavy elements had been suggested much earlier[10]. However, at the time of the Lysekil conference it had also been suggested that the next magic proton number could be Z=114[11], not Z=126. These developments led to an explosion of interest in superheavy elements and large theoretical and experimental efforts followed. Some of the first extensive theoretical studies were the ones with Szymanski and his colleagues[2,3,4]. For one of many reviews of the theoretical activities see Nix[12]. Experimental efforts continued to be negative through the 1970ies[13].

However, none of these early efforts led to very firm conclusions because different models and corresponding calculations gave very different results. Experience has now taught us that to obtain models that could, with some confidence, predict properties of unknown nuclei it was essential to 1) further develop key aspects of the models and 2) *systematically* apply the models to *large* regions of the periodic chart to determine model constants such as spin-orbit strengths and the constants of the macroscopic part of the model. For models of the macroscopic-microscopic type many such developments took place in Los Alamos during the first few years of the 1970ies.

2. 1970–1990: The Maturing of the Macroscopic-Microscopic Model

In Los Alamos Ray Nix and collaborators developed a macroscopic-microscopic model based on on the liquid-drop[14] macroscopic model and the folded Yukawa single-particle model[15]. It was almost immediately applied to a calculation[16] of superheavy element decay properties, which is widely cited. However, several developments soon after this investigation led to significant enhancements in the model. They were 1) a new set of spin-orbit strength and potential diffuseness parameters, 2) inclusion as options additional shape parameterizations that are better suited for studying nuclear ground-state shapes, and 3) taking into account finite-range effects in the nuclear force, leading to a new description of the surface energy in the macroscopic part of the model.

The first two developments were the result of a focussed collaboration between Ray Nix, Peter Möller, who as a graduate student visited Los Alamos during all of 1973, and his thesis advisor Sven-Gösta Nilsson who was on sabbatical leave at Los Alamos during the first half of 1973. In the original implementation of the folded-Yukawa model the single-particle spin-orbit and diffuseness parameters were obtained by optimizing the calculated single-particle level spectra to the observed single-particle levels in ^{208}Pb[15]. Sven-Gösta Nilsson observed that with these parameters levels in deformed rare earth and actinide nuclei were poorly reproduced. By comparing calculated and experimental single-particle levels in both deformed and spherical nuclei from the rare-earth region through the actinide region new spin-orbit and diffuseness parameters were determined, different sets for the rare-earth and actinide regions. Also crucial in this study were optimum ground-state shapes, which we could only access in the new parameterization, the Nilsson perturbed-spheroid ϵ parameterization, that we implemented in the associated computer codes. In a two-dimensional space spanned by ϵ_2 and ϵ_4 it was found that this parameterization usually resulted in lower ground-state energies than a two-dimensional constrained version of the three-quadratic-surface parameterization, by up to 1.2 MeV for ^{232}Th with the droplet model macroscopic energy[17]. These developments are discussed in[18,19]. Subsequently, it was postulated that the spin-orbit strength varied linearly with nucleon number A. Therefore the values determined in the rare earth and actinide regions[19] completely define the spin-orbit strength throughout the nuclear chart[20]. When computers became sufficiently powerful to allow systematic, global studies it was shown that this choice of spin-orbit strength and the new diffuseness constant gave excellent agreement between calculated and experimental ground-state spins, see for example Figs. 2 and 3 in[21] and the slightly refined calculation in Figs. 17 and 18 in[22].

The third development at this time was a result a joint effort between Los Alamos and outside collaborators, namely the formulation of a modified description of the surface energy in the liquid-drop model, sometimes referred to as “finite-range effects of the nuclear force”[23]. The model was subsequently further refined[24]. Macroscopic-microscopic studies incorporating the initial formulation are in[19]. At this time also the droplet model of Myers and Swiatecki was sometimes used as the macroscopic model[18,17,19].

In parallel with these developments, the first macroscopic-microscopic global nuclear mass calculation with *calculated* shell corrections was performed, also at Los Alamos[25,26]. Because computers were not very powerful at this time these calculations were based on the less computer-intensive Nilsson modified-oscillator single-particle potential to calculate the microscopic shell corrections. However, the results were encouraging, with a 0.70 MeV rms deviation with respect to known nuclei. Later it could be shown that it had much better predictive power than models with postulated shell corrections[27].

Several years after these developments Treiner and PM who both spent sabbaticals 1983–1984 at LBL, embarked together with Myers and Swiatecki on an effort to understand and address known issues in the droplet model, for example the tendency of its parameters to take on unphysical values and the excessive softness of the mass surface with respect to neutron excess. The initial approach was to combine the droplet model with the folding-model Coulomb and surface-energy integrals. However, there were still difficulties because the droplet-model first-order expansion of the macroscopic energy in terms of $A^{-1/3}$ and neutron excess $I = (N-Z)/(N+Z)$ did not describe compressibility effects sufficiently accurately. The expansion started to break down already at $A \approx 100$. To address this issue a somewhat empirical exponential term was incorporated in the energy expression. Details are in [28,29]. At the time we were unable to carry out a full-fledged global mass calculation in this model but presented a mass table in which effects of the ϵ_3 and ϵ_6 shape degrees of freedom were determined by an interpolation scheme based on studies of only a few nuclei. Interestingly we obtained a global rms deviation of 0.676 MeV, surprisingly close to 0.673 MeV (table D in[30]) rms deviation of the current published mass model version (FRDM (1992)), when both are adjusted to the 1977 experimental data set[31].

With all these model refinements incorporated over more than a decade we had to wait for substantial improvements in computer power before the enhanced models could be fully explored. An hint of what might come was the 1981 mass calculation[20,32].

3. 1990–Present: Large-Scale Computing Based on Mature Models

Already in 1981 we presented our first global mass calculation in a macroscopic-microscopic model that included most of the above features[20,32]. After some discussion PM and Nix decided not carry the calculations to the drip lines nor into the superheavy-element region, but only include nuclei about four nucleons beyond stability so that the model could be further tested against subsequently discovered nuclei before the calculations were extended to the drip lines and into the superheavy region. In hindsight this was an unfortunate decision on the side of caution. The model turned out to have excellent predictive capabilities[33]. Those heavy elements that were included showed very strong shell effects (about -7 MeV) near $Z = 108$ and $N = 162$, as could also been seen in the earlier tables of[17]. The discoveries, soon after the publication of this mass table, of elements $Z = 107$, 108, and 109 at GSI led us to extend the calculations into the superheavy region[34]. Our most current, published mass table is the FRDM(1992) finalized in 1992 was published in 1995. These types of calculations also, often automatically, provide many other ground-state properties of nuclei, such as moments and spins, this is well-known, see for example[22]. We are now in the process of further improving the accuracy of our mass calculations and have achieved a model accuracy of 0.596 MeV. The details are in[35]. We refer to this (interim) table as FRDM(2007b). Rather than publishing a long string of mass tables corresponding to each feature we investigate we plan to investigate a few additional possible enhancements before we present a new FRDM "edition". We compare in Fig. 1 calculated masses to experimental data.

What we have not fully known in terms of firm numbers is: how important were the several enhancements added to the model over the years. Specifically, how important were the new spin-orbit parameters introduced in 1973 for global mass calculations? We have now for the first time carried out such a study. We perform a full-fledged mass calculation following exactly the same steps as in FRDM(2007b), except we use the initial spin-orbit and diffuseness strengths from[15]. We compare in Fig. 2 those calculated masses to experimental data. We note that the mass model error with the old 1970 single-particle parameters is 0.691 MeV which is substantially higher than the value 0.596 MeV which we obtain with the redetermined

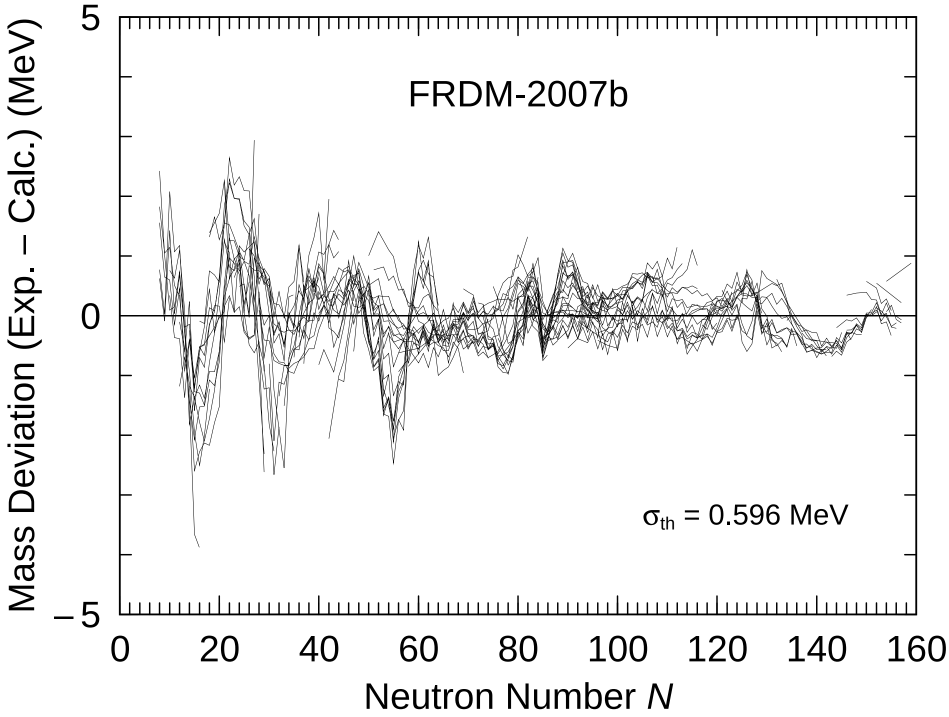

Fig. 1. Differences between masses calculated in the FRDM and experimental masses [36]. Above about $N = 65$ the model error is only 0.34 MeV. In the light region the error is much larger. The regularity of the error fluctuations suggests it might be possible to find a model enhancement to substantially reduce them. Another possibility is the model gradually deteriorates as the nucleon number decreases.

single-particle parameters. This is somewhat remarkable, because at the time the parameters were determined *no* consideration of masses affected the choice, only the spins of odd-even nuclei were considered. Obviously, in a perfect model you only need n data points to determine n model parameters. However, nuclear-structure models are not perfect, they are based on imperfect effective interactions and imperfect models. But our result here shows that the model has remarkable internal consistency: a parameter choice that improves the single-particle spectrum also substantially improves other model properties.

It is well known that most early theoretical studies of the heavy-element region did not identify the region of *deformed* elements connecting the actinide region with the superheavy island. One reason is most studies[3,16] did not look at this region, because at the time it was assumed these nuclei would be highly unstable. One exception is one of the first studies in the folded-Yukawa model with the new single-particle parameters: in[19]. Here the tabulated shell correction for the ground-state of ^{270}Hs is -8.05 MeV, for example. Another study of stability in the heavy-element

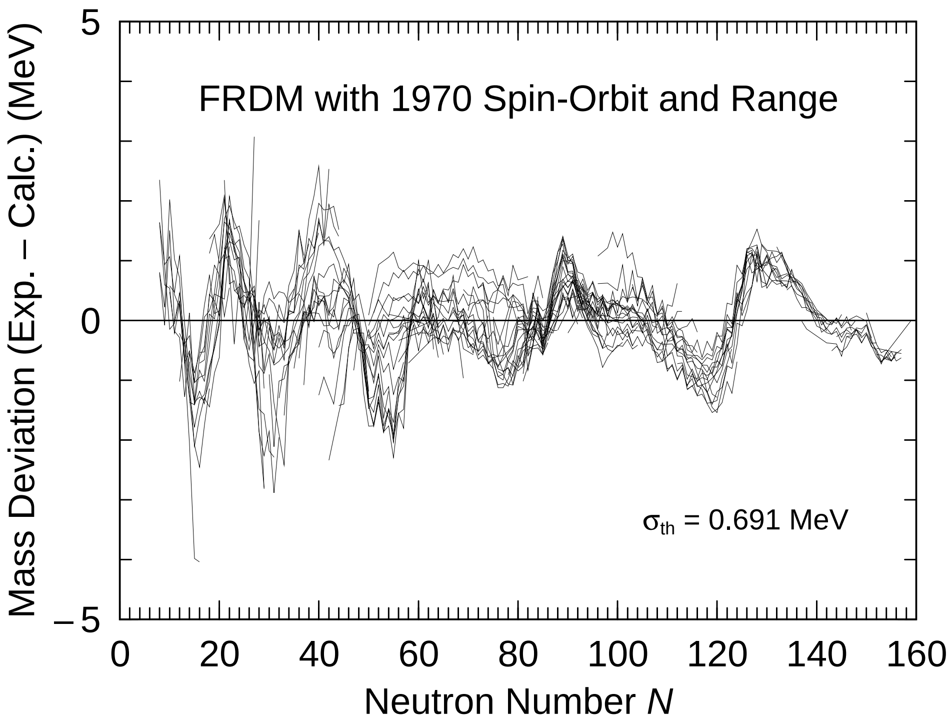

Fig. 2. Differences between masses calculated in the FRDM and experimental masses [36]. The spin-orbit strength and potential diffuseness are those originally used in the folded-Yukawa single-particle potential. They were determined by fitting calculated single-particle spectra to experimental levels in nuclei near ^{208}Pb. However, this optimization to a limited region yields much larger deviations in the heavy region than is obtained with parameters determined from comparing to a larger, global data set.

region[37,38], does use the new single-particle parameters, but surprisingly there is no local fission-barrier-height maximum at $Z = 108$ and $N = 162$. This would have been expected if the calculated ground-state shell corrections had been large and negative, see for example[39], where such a local maximum is clearly present. We feel that the reason for the non-observance is that the shapes studied did not include shapes with large negative values of β_4 which are necessary for the large negative shell corrections to appear. In[37,38] the full three-quadratic-surface shape parameterization was restricted to a two-dimensional space.

To further investigate how sensitive the shell stabilization of the region near $Z = 108$ and $N = 162$ is to model assumptions we plot in Fig. 3 the shell corrections we obtained in our full-fledged mass calculations with the single-particle parameter sets used in our current model, corresponding to Fig. 1, and in Fig. 4 the shell corrections corresponding to the old parameters. The two calculations are very similar in structure, although absolute values can differ by an MeV or more. In

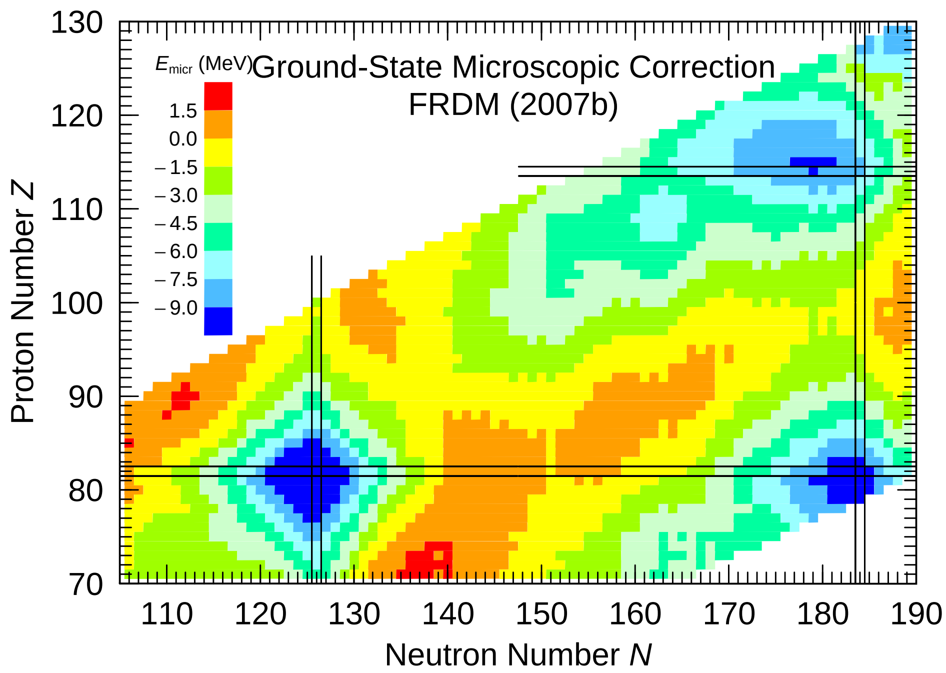

Fig. 3. Microscopic shell corrections calculated in our current interim FRDM(2007b). They are very similar to the results of FRDM(1992) [30], but the mass table corresponding to FRDM(2007b) agrees better with data than does FRDM(1995).

particular, also with the 1970 choice of spin-orbit strength we find enhanced stability near $Z = 108$ and $N = 162$. Since similar results are obtained in macroscopic-microscopic calculations based on a Woods-Saxon single-particle potential[40], we can say that the theoretical result that nuclei near $Z = 108$ and $N = 162$ are relatively stable is very robust. It is unfortunate that all these detailed results were not available *before* the first experimental observations in this region. Had they been, a robust *prediction* of the existence of a totally unexpected region of stability could have been made.

The results in Figs. 1 and 2 show that optimized single particle parameters can improve a mass model accuracy by at least 0.1 MeV or, in this case by 14%. We know from previous results that also the type of macroscopic model is important. In[30] results based on a generalized droplet model were 0.12 MeV or 14% more accurate than result based on a generalized liquid drop model. The droplet-based results also exhibit other desirable features. In Fig. 5 we compare both mass calculations to new masses that were not included in the adjustment of the model parameters; in most cases they were not even measured at the time. The FRDM (1992) agrees *better* with the new data (0.462 MeV) than with the masses to which it was adjusted

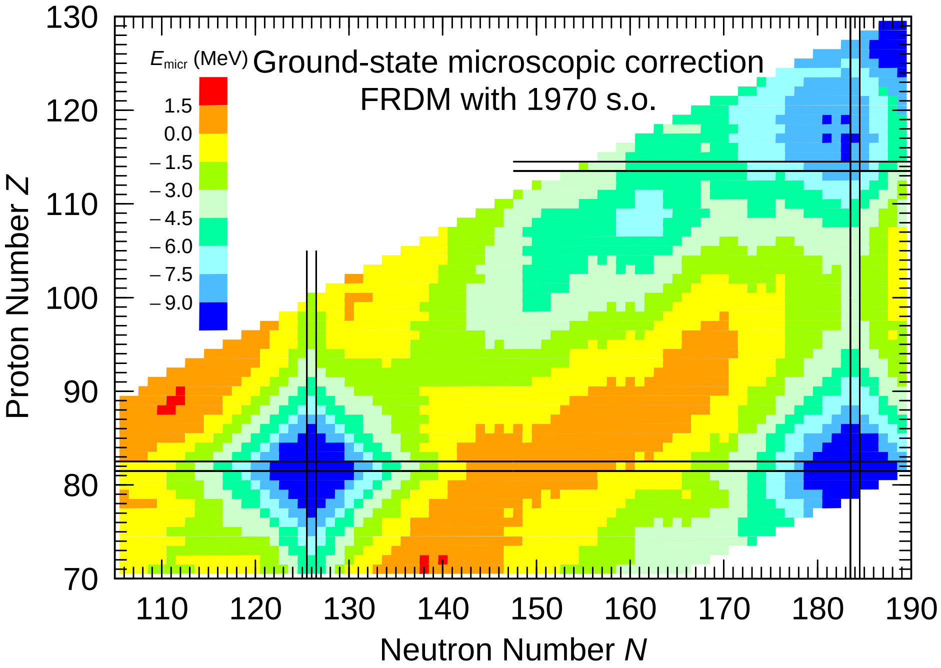

Fig. 4. Microscopic shell corrections calculated in our FRDM model but the spin-orbit and diffuseness parameters of the folded-Yukawa single-particle potential are different than our current choice, they have the values originally used [15]. The shell corrections are different in value than with our current parameters, but the structure of the microscopic correction is very similar to Fig. 3, but the center of the superheavy island is closer to $N = 184$.

(0.669 MeV). The FRLDM (1992) with its generalized liquid-drop macroscopic model agrees about equally well with the data set to which it was adjusted and the newly measured masses. However, it also has a systematic trend: calculated binding energies grow increasingly too small with increasing proton excess. This trend is entirely absent in the FRDM results. The good agreement and absence of systematic deviations indicate that effects like compressibility and redistribution are well described within this macroscopic model. In the liquid-drop model, shown in the top plot, charge cannot redistribute towards the surface to increase the binding energy. This results in too low binding energies, in particular for proton-rich nuclei.

4. After a Long Journey: Arriving at the Shores of the Superheavy Island

In the 1960–1970 time frame a common belief was that the super-heavy "island" was isolated from the peninsula of known elements by a sea of instability. But, starting in 1981, the discoveries of the elements from $Z = 107$ to $Z = 112$ at GSI[41,42,43]

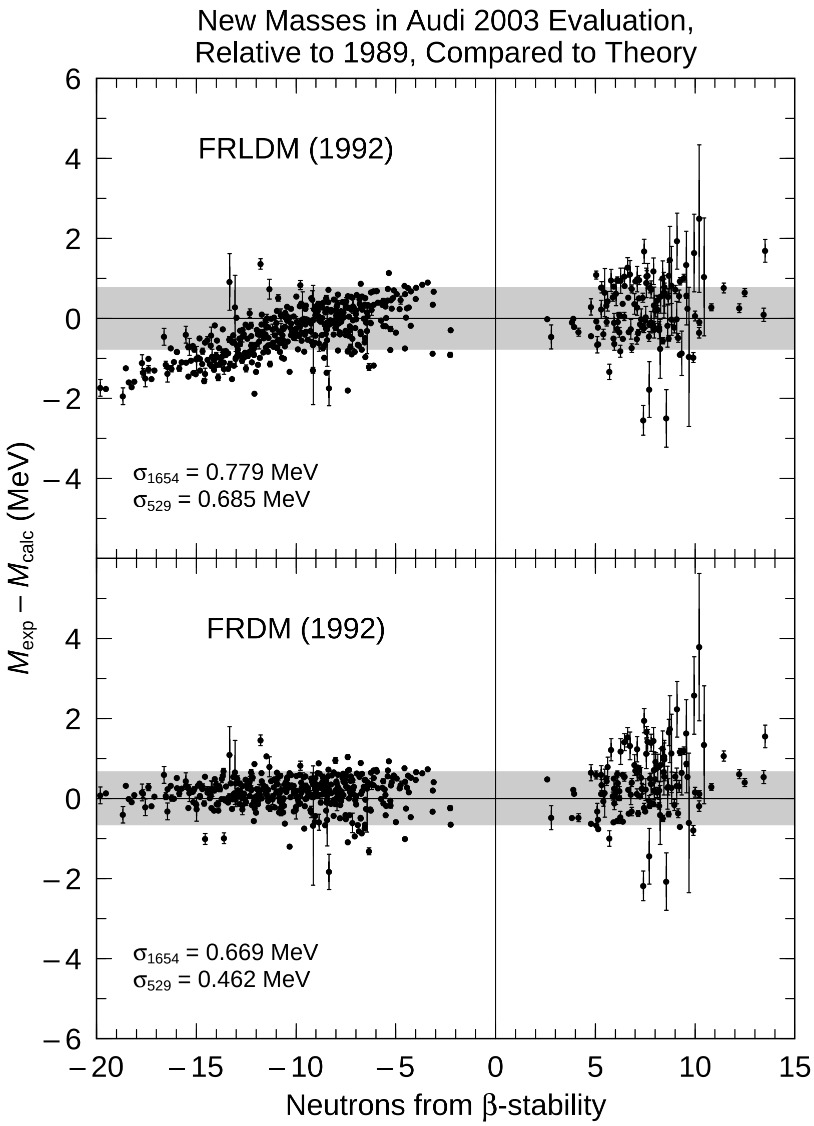

Fig. 5. FRDM and FRLDM compared to new mass measurement data which were not available when the model constants were determined in 1992. The FRDM agrees *better* with the new data (0.462 MeV) than with the mass data to which it was adjusted (0.669 MeV). The FRDM is well-behaved far from stability, the FRLDM shows systematic deviations, see text for discussion.

showed that the peninsula of known elements is connected to the doubly-magic superheavy island by a relatively narrow land bridge. Although these nuclei are deformed in their ground states they are highly stabilized with respect to fission by large gaps in the *deformed* level spectra, most notably $Z = 108$ and $N = 162$.

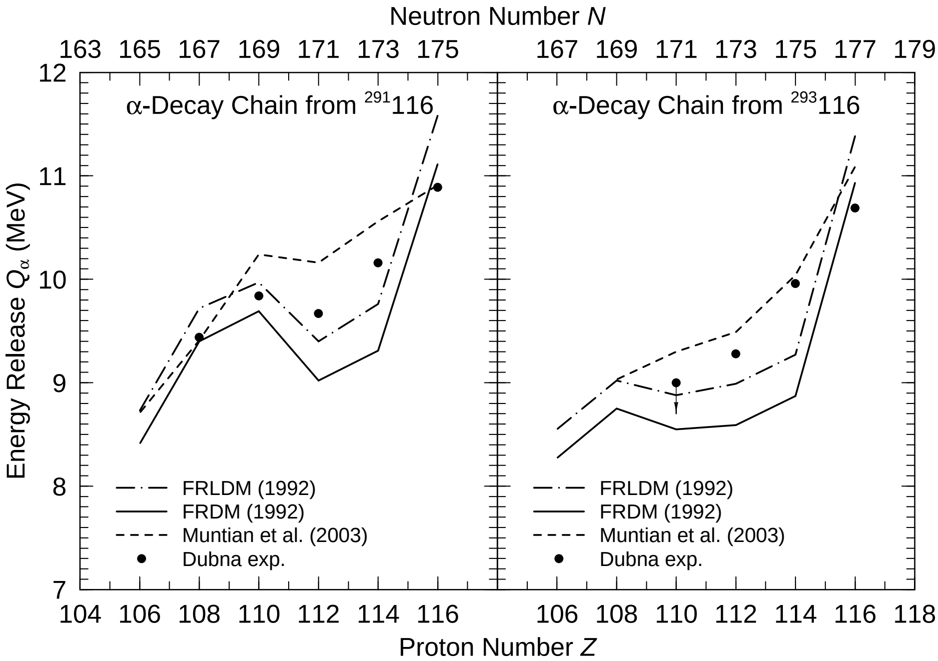

Fig. 6. Comparison between two observed α-decay chains and three models. The FRDM has a kink at $Z = 114$ which is absent in the experimental data. The features of the $^{291}116$ decay chain are reasonable well reproduced, in particular if we recall that this chain is thirty nucleons heavier than the heaviest nucleus used in the adjustment of model parameters.

Some calculations agree well with the observed structure seen in the Q_α energies in the decay chains[22,43].

But let us focus here on the more recent studies of reactions leading to compound systems with proton numbers reaching almost $Z = 120$, which have mainly been undertaken at Dubna[44]. Since most of the α-decay chains terminate by fission rather than reaching known nuclei, identification and confirmation has not been straightforward. However, recently several confirming experiments have been performed, for example[45,46,47] for some of these reactions. We compare in Fig. 6 calculated Q_α to the Dubna data for two chains. Clearly the data have no kink at the supposedly magic proton number $Z = 114$. This could mean a) that $Z = 114$ is not magic or b) that the magicity is not apparent this far from $N = 184$. The data has a clear kink at $Z = 112$, which is also present to varying degrees in the three models plotted. Some constants in the Muntian macroscopic model have been adjusted to some Q_α data in the heavy element region[48,49], in the FRDM/FRLDM they have not.

An important issue in studies of the superheavy island is reaction mechanisms.

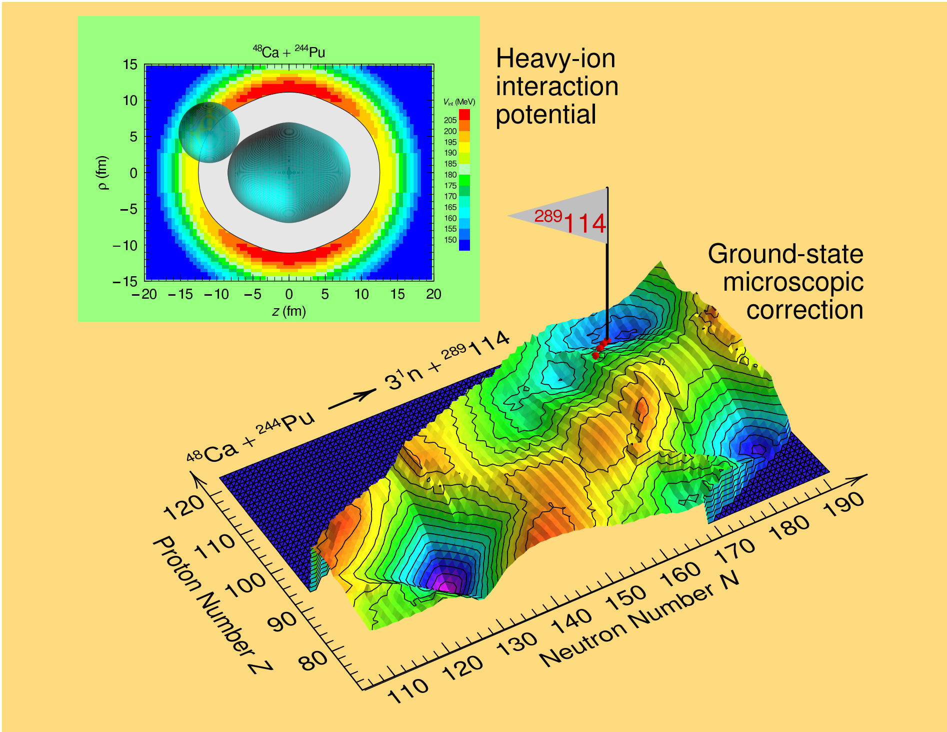

Fig. 7. Calculated shell corrections in the heavy-element region. Superimposed is the location of a decay chain observed at Dubna. The insert is a plot of the calculated two-dimensional fusion barrier between the deformed target and spherical projectile, see text for further comments.

Many reaction models are in terms of spherical models of target and projectile. In our opinion this is inadequate for hot-fusion heavy-ion reactions, since in these reactions the target is an actinide nucleus, which is well deformed. In Fig. 7 we show the shell correction calculated in the FRDM(1992) model with one of the Dubna decay chains superimposed on the calculated shell correction. We also show, as an insert the calculated potential between a projectile and target for the reaction ^{48}Ca+^{244}Pu. The energy in each point corresponds to the potential energy between projectile and target when the center of the projectile is in that (z, ρ) location. There is a substantial difference in the barrier height in the polar and equatorial directions. A projectile incident in the equatorial direction will need a 15 MeV higher energy to reach the ("Coulomb") barrier than in collisions in the polar direction. Lower compound-system excitation energies have been considered advantageous for forming evaporation residues, so in early hot fusion experiments energies appropriate for the barrier in the polar direction were preferred[50]. In a 1994 calculation of heavy-ion fusion barriers between deformed nuclei, we pointed out that higher energies would allow penetration of the equatorial Coulomb barrier. The more compact, spherical-like touching configuration would be more nearly "inside" the fission saddle point of the compound system than a polar-collision configuration. This could more than well compensate for the higher compound-system excitation energy and increase the cross section for evaporation-residue formation. It was suggested that only a small "extra push" might be needed in equatorial collisions to take the trajectory inside the fission saddle point, a necessary condition for compound nucleus formation[51].

5. Summary

I have given a snapshot of developments I experienced together with my mentors, that were important in superheavy element research (a very recent more general review is in Ref.[52]). To understand the significance of some of the developments we studied the accuracy of FRDM macroscopic-microscopic mass calculation with two substantially different choices of single-particle parameters, each calculation with optimized macroscopic-model parameters. We believe that it is the first time two single-particle parameter choices have been so directly and consistently compared. We learned that

- Adjusting the spin-orbit and the diffuseness parameters so as to optimize the agreement between calculated and experimentally measured spins of odd-A nuclei have an enormous effect on mass model accuracy; it improved from 0.691 MeV to 0.596 MeV, a gigantic improvement when the model deviations from data are so low already. This is all the more remarkable, since masses were not considered at all in this optimization of single-particle parameters.
- The shell corrections in the heavy-element region are very similar with the two single-particle parameter choices. In particular we find in both results a region of substantial shell corrections connecting the actinide region with the spherical superheavy region. Since similar results are obtained with a universal-parameter Woods-Saxon model we can say these results are very robust. It is of course now known that these results are in good agreement with experimental observations on these nuclei.

The Dubna hot-fusion experiments have accessed several new elements beyond those reached in cold fusion. The range of nuclei observed hint at a new continent, rather than an island in this region. It is exciting to anticipate further developments in experiment and theory that will lead to more complete understanding of both the properties of the nuclei in this region and the reaction mechanisms that will allow us to map the extension of the continent.

6. Dedication

I learned with great sadness, just after this conference, that the last of my four principal mentors, Wladek Swiatecki passed away September 30, 2009 to join my other three mentors. At the same time I can only feel great joy over the incredible scientific journey they guided me on during my professional career and dedicate this contribution to my mentors Sven-Gösta Nilsson, Ray Nix, Wladek Swiatecki, and Zdzislaw Szymanski.

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