# **A timeline of the important theoretical developments leading to our current insights into heavy-element stability**

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Collaborators on this and other projects,

see coauthors on papers posted on URL below and publications listed at end.

More details about masses, other projects (beta-decay,fission), associated ASCII data files and figures are at

# **http://t2.lanl.gov/nis/molleretal/**

and at list of relevant publications after conclusions.

Stanislav Ulam has remarked: It is remarkable how a few characters scribbled on a blackboard can change the course of world history.

Feynman:

- I do not care how smart you are
- or how complicated your model is
- If it does not agree with experimental measurements it is wrong!

# **Nuclear BINDING ENERGY Bethe-Bacher (-Weizacker) (1936) ¨**

$$
B(N, Z) =
$$
  
\n
$$
+a_V A
$$
 (Volume energy)  
\n
$$
-a_S A^{2/3}
$$
 (Surface energy)  
\n
$$
-a_C \frac{Z^2}{A^{1/3}}
$$
 (Coulomb energy)  
\n
$$
-a_I \frac{(N-Z)^2}{A}
$$
 (Symmetry energy)  
\n
$$
-\delta(A)
$$
 (Pairing energy)

Bethe and Bacher, Revs. Mod. Phys. **8** (1936) 82 1

Bethe and Bacher, Revs. Mod. Phys. **8** (1936) 82 ( in §33):

"There remains thus the nucleus containing 8 neutrons and 8 protons, i.e.,  $16$ O, to test the shell structure" hypothesis by means of nuclear energies. It seems in fact that there is ample evidence for a particular stability of  $16$ O, and thus for the individualparticle approximation."

So, already in 1936, shell-structure, single-particle models, and how they might modify a macroscopic model were in mainstream discussions.

Hahn and Strassman conclusively identified barium in the products after bombarding uranium with neutrons

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(Naturwiss. 27 (1939) 11)
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Meitner and Frisch proposed that observa-

tions of barium in the reaction products were

due to nucleus deforming like a drop

(Nature **143** (1939) 239)

Frisch measured (the predicted) fragment high

kinetic energies

(Nature **143** (1939) 239)

Bohr and Wheeler Calculated

(Phys. Rev. **56** (1939) 426):

# **Nuclear POTENTIAL ENERGY**

**versus deformation**

# $B(N,Z) =$  $+a_VA$  (Volume energy)  $-a_S A^{2/3} B_s(\beta)$  (Surface energy)  $-a_{\mathsf{C}}$  $Z^2$  $A^{1/3}$  $B_C(\beta)$  (Coulomb energy)  $-a_I$  $(N - Z)^2$ A (Symmetry energy)  $-\delta(A)$  (Pairing energy)

Swiatecki (and others) observed that experimental actinide spontaneous-fission half-lives differed substantially from what could be explained from smoothly varying (with neutron number and proton number) liquid drop barriers.

He correlated the differences with differences between liquid-drop ground state masses and measured masses and found that such ground-state "shell structure" could account for the observed behavior of actinide spontaneous fission half-lives.

trend is consistent with a straight line, defining  $(Z^2/A)_c$  $=40.2\pm0.7$ . The equation of the line leads to the semiempirical formula,

$$
M_2 - M_1 = 0.090(40.2 \pm 0.7 - Z^2/A)^{\frac{1}{2}}A. \tag{4}
$$

One may combine Eq. (4) with the relation:

$$
M_2+M_1=A-\nu
$$

 $(\nu =$  number of neutrons emitted in fission),

to predict the positions of the peaks in the yield curves of elements that have not yet been investigated. If an average value  $\bar{\nu} = 2.8$  is used, one finds

$$
M_2 = \frac{1}{2}A - 1.4 + 0.045(40.2 \pm 0.7 - Z^2/A)^2 A, \quad (5)
$$

$$
M_1 = \frac{1}{2}A - 1.4 - 0.045(40.2 \pm 0.7 - Z^2/A)^{\frac{1}{2}}A. \tag{6}
$$

The present analysis provides a reason for the empirical observation that in the fission of different elements the position of the heavy peak remains

TABLE I. Positions of the peaks in the fission yield curves.

Position of peaks				,	
Observed <sup>®</sup>		Formulas $(5)$ , $(6)$			
M <sub>2</sub>	$M_1$	M2	$M_1$	Remarks	Reference
140	91	139.1	91.1)		b
140	98	141.1	95.1	Low-energy	c
138.5	95	138.2	95.0	neutron	d
137	93	136.2	95.0	fission	b, e
138	99	137.9	99.3		c
140	96	140.2	95.0		f, g
136	103	134.7	104.5		
139	108	140.2	109.0		$_{\rm h}^{\rm g}$
					Spontaneous fission

**The uncertainty in the observed values of**  $M_2$  **and**  $M_1$  **is of the order<br>of**  $\pm 1$  **or**  $\pm 2$  **mass units. (It is more in the cases of U<sup>398</sup> and U<sup>388</sup>, No<br>systematic attempt has been made to adjust**  $A-M_2-M_1$  **to agree wi** 

approximately constant. If the degree of asymmetry remained unchanged from nucleus to nucleus, both peaks would move towards higher masses with increasing  $A$ . In fact, there is superimposed on this shift a coming together of the peaks with increasing  $Z^2/A$ . Since the over-all trend of  $Z^2/A$  is to increase with A, the result is that for the light peak the two shifts add up whereas for the heavy peak they partly cancel. This is illustrated in Table I, where  $M_2$  and  $M_1$ , calculated according to (5) and (6), are compared with the observed values.

Further measurements of fission asymmetries would be interesting, especially in the region of  $Z^2/A$  close to the critical value, where the present considerations suggest a rapid decrease of  $M_2-M_1$ .

It is a pleasure to acknowledge stimulating discussions with Professor S. G. Thompson, Dr. A. C. Pappas, and Dr. T. Maris.

<sup>1</sup> N. Bohr and J. A. Wheeler, Phys. Rev. 56, 426 (1939).<br>
<sup>2</sup> A. E. S. Green, Phys. Rev. 95, 1006 (1954).<br>
<sup>3</sup> W. J. Swiatecki (to be published).<br>
<sup>4</sup> D. L. Hill and J. A. Wheeler, Phys. Rev. 89, 1102 (1953).

#### **Systematics of Spontaneous Fission** Half-Lives

W. J. SWIATECKI

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SEVERAL authors have noted the over-all trend of<br>spontaneous fission half-lives to decrease with increasing  $Z^2/A$  as well as the considerable deviations (by several powers of 10) from any smooth dependence on this parameter.<sup>1</sup> We should like to discuss the close correlation which seems to exist between the half-lives and the finer details in the systematics of the groundstate masses of nuclei.<sup>2</sup>

A simple way of exhibiting this correlation is to plot the deviation  $\delta \tau$  from a straight line in a plot of  $\tau$ [ $\tau$ =log<sub>10</sub>(half-life)] vs  $Z^2/A$ , against deviations ( $\delta M$ ) of the masses  $M$  of the nuclei from a smooth reference surface  $M_{ref}(A,Z)$ . We made such a plot, with  $M_{ref}$ taken to be the semiempirical mass surface of Green<sup>3</sup> (based on the liquid drop model):

$$
\delta M = M - M_{\text{ref}},
$$
  
\n
$$
M_{\text{ref}} = 1000A - 8.3557A + 19.120A^3 + 0.76278Z^2/A^3 + 25.444(N - Z)^2/A + 0.420(N - Z) \text{ millimass units.}
$$
 (1)

The experimental masses  $M$  were taken from Glass  $et~al.^4$ 

In the case of even-even nuclei the plot of  $\delta\tau$  vs  $\delta M$ suggested a series of straight lines, one for each Z. indicating that for the isotopes of one element special stability of a nucleus (small  $\delta M$ ) is invariably associated with a longer lifetime (large  $\delta \tau$ ). The lines had approximately the same slope, thus defining a spontaneousfission hindrance factor which corresponds to about  $10<sup>5</sup>$  times longer lifetime for each millimass unit of extra stability. This suggested that if the observed lifetimes were corrected for the variations in stability of the ground states, a more regular dependence of  $\tau$ on  $Z^2/A$  might be discernible.

Figure 1 shows the effect on the plot of  $\tau$  vs  $Z^2/A$  of adding to the observed  $\tau_{\rm exp}$  an empirical correction  $k\delta M$  ( $k\sim$ 5 if  $\delta M$  in mMU). For even-even nuclei the values of  $\tau_{\exp} + k \delta M$  define a fairly smooth curve, with indications of a similar curve for odd-A nuclei. [In a preliminary plot the hindrance factor  $k$  was taken to be 5. A small but significant further smoothing of the points resulted from making k vary with  $Z^2/A$  according to  $k=5-(Z^2/A-37.5)$ . This is the case shown in Fig.  $1.7$ 

The result can be stated in the form of an empirical formula for half-lives; e.g., for even-even nuclei,

$$
\tau_{ee} = f(Z^2/A) - k\delta M,\tag{2}
$$

where  $f$  is the curve defined by the even-even points in Fig. 1. The relation of the points for odd- $A$  nuclei to the curve obtained from  $(2)$  by a shift upwards of 6.6 units is also shown in Fig. 1. The lifetime of the odd-odd nucleus E<sup>254</sup> (einsteinium,  $Z=99$ ) is consistent with a further shift of 4.9 units. The curve  $f(Z^2/A)$  can be



FIG. 1. Plot of spontaneous fission half-lives against  $Z^2/A$ . The Figure ; the of spannatous instant part of the figure; the "corrected"? values  $\tau_{exp} + k\delta M$  group themselves around the figure; the "corrected"? values  $\tau_{exp} + k\delta M$  group themselves around the three curves. Experimental p symbols which, reading from left to right along the odd-A curve,<br>refer to U<sup>235</sup>, Pu<sup>239</sup>, Bk<sup>249</sup>, Cf<sup>249</sup>, E<sup>255</sup> (einsteinium, Z=99), and<br> $\text{Fm}^{255}$  (fermium, Z=100). The odd-odd nucleus E<sup>254</sup> is marked by a square.

represented for example by a cubic, which leads to the following formulas for the lifetimes:

$$
\tau_{ee} = 18.2
$$
  
\n
$$
\tau_{\text{odd }A} = 24.8
$$
  
\n
$$
\tau_{oo} = 29.7
$$
  
\n
$$
\tau_{00} = 29.7
$$

where  $\theta = (Z^2/A) - 37.5$ , and  $\delta M$  is the deviation in mMU of the experimental mass from the surface (1). Table I compares the observed half-lives with the values calculated by means of (3). The remarkable

	Nucleus	Experi- mental <sup>a</sup>	Formula (3)	Nucleus	Experi- mentala	Formula (3)	
Even-even nuclei				Even-even nuclei			
Th U Pu	-230 232 232 234 236 238 236 238 240 242 244 Cm 240 242 244 246	$\geqslant7.18$ 18.15 13.90 16.30 16.30 15.90 9.54 10.69 11.08 10.86 10.40 6.28 6.86 7.15 7.48	19.39 18.84 13.56 15.98 15.21 15.52 9.66 11.57 11.09 11.22 10.13 6.27 7.27 7.09 7.88	Сf 246 248 250 252 254 Fm 254 256 U 235 Pu 239 Bk 249 Сf 249 Е 253 Fm 255	3.32 3.85 4.18 1.82 $-0.70$ $-0.30$ $-3.52$ Odd-A nuclei 17.26? 15.74 8.78 9.18 5.48 1.30 Odd-odd nuclei	3.27 3.92 4.24 1.60 $-1.02$ $-0.85$ $-3.02$ 18.02 15.42 8.67 8.65 4.38 2.79	
				Е 254	5.18	5.17	

TABLE I. Values of log<sub>10</sub>(half-life).

<sup>a</sup> The experimental values are from a summary by A. Ghiorso, kindly lent to me by Professor S. G. Thompson.

degree of smoothing achieved by means of the unsophisticated correction  $k\delta M$  is illustrated by the fact that the deviations from  $(3)$  rarely exceed 0.5. (Note that a shift in  $\tau$  of this amount would be produced by an error of 0.1 mMU in  $\delta M$ .)

The importance of shell structure in the fission process is suggested by the fact that, according to the present considerations, the oscillations of the masses (associated with individual particle structure) in the range  $\delta M = 1-3$  mMU shorten the lifetimes by factors of  $10^5$  to  $10^{15}$ . On the other hand the irregularities in the original plot of  $\tau_{\exp}$  against  $Z^2/A$  are seen to be largely due to irregularities in the ground-state masses, associated with shell structure in the ground-state configuration. The smoothness of the points  $\tau_{\exp} + k \delta M$ suggests that, after correcting for shell structure in the ground-state configuration, the description of the fission process in terms of a model in which singleparticle features are treated in an average way may be useful. Qualitative reasons for the greater validity of such an averaged description for the more strongly deformed nuclear shapes occurring in fission may be found in the disappearance for such shapes of degeneracies in the energy spectrum associated with the proximity to a spherically symmetric configuration.

It is a pleasure to acknowledge discussions with Professor S. G. Thompson and Dr. A. C. Pappas and stimulating contacts with Dr. Aage Bohr and Dr. B. R. Mottelson and members of the C.E.R.N. Theoretical Study Group in Copenhagen.

<sup>1</sup> See for example J. R. Huizenga, Phys. Rev. 94, 158 (1954). <sup>2</sup> The existence of correlations between nuclear masses, fission thresholds, and half-lives has been considered by Professor D.

Erisch, to whom I am greatly indebted for stimulating discussions.<br>Fisch, to whom I am greatly indebted for stimulating discussions.<br> $^{8}$  A. E. S. Green, Phys. Rev. 95, 1006 (1954). <sup>4</sup> Glass, Thompson, and Seaborg, J. Inorg. Nuc. Chem. 1,

3 (1955).

Are the heaviest actinides ending the periodic system of observable elements? Scharff-Goldhaber mentions in Nucleonica already in 1957 that *There may be for instance, another region of relative stability at the doubly-magic nucleus* <sup>126</sup>*X* <sup>310</sup> *(the closing of the j neutron shell)"*

Since she mentions it so casually this possibility was probably always recognized by the community. Exactly which nuclei might be stable was overlooked in calculations for a further 10–25 years, see below.

# nuclear physics

#### By GERTRUDE SCHARFF-GOLDHABER

EVEN A SUPERFICIAL GLANCE backward will teach us that it is impossible to predict in detail the future of a fastmoving science like nuclear physics. It will remind us that often entirely unexpected events changed the direction of endeavor in this field. These events were either of an experimental nature, as, for instance, the finding that beta rays have continuous energy spectra, or they consisted in the formation of new concepts—as, for example, of the liquid-drop model of the nucleus.

It is, of course, similarly impossible to predict what extraneous happenings may in the future affect scientific progress as profoundly as two world wars and political persecution have affected it in the past. During the last twelve years the great importance attached to atomic energy has induced an unprecedented increase in the tempo of research, and new nuclear physics centers have sprung up all over the world. This development is viewed by many with delight while others are afraid that it may have a negative effect on nuclear physics as a pure science.

In spite of all the uncertainties mentioned, it is useful to interrupt from time to time one's preoccupation with the problem at hand to investigate the trends that current research seems to follow, both in experiment and theory, and to try to recognize how far these may serve to bring us closer to the solution of outstanding problems.

The central problem is to understand the nucleus in the same sense in which one might have said in 1926 that the atom was understood: one knew then not only that the forces between the nucleus and the atomic electrons were pure Coulomb forces, but also that the excited states in which the system could exist were governed by the laws of quantum mechanics, including the Pauli exclusion principle. (It is true, only the states of the simplest atom, hydrogen, could be exactly calculated, while already the helium atom presented such overwhelming mathematical difficulties that it took about another quarter century and development of computers to reach the same stage.)



If we want to study events happening inside the nucleus, its finite extension and the distribution of charge and current inside it, we have a task of a higher degree of difficulty than in the atomic case: (a) We do not know the exact nature of the force between two nucleons, nor do we know whether a potential exists for this force. (b) One cannot consider, in first approximation, the interaction of two nucleons only, because all nucleons are close to each other. (c) We do not know whether the laws of quantum mechanics are sufficient to describe a nucleus completely.

#### Quantum mechanics may need to be modified, e.g., by the introduction of the concept of a fundamental length, before nuclear phenomena can be explained

On the other hand, we have two important clues on the nature of nuclear forces:

1. Apart from the lightest ones, all nuclei have the same density  $\rho = 1.7 \times 10^{38}$  nucleons/cm<sup>3</sup>, at least in their central part, so that it is reasonable to speak of "nuclear matter." Hence, if it were not for the Coulomb repulsion between the protons, nuclei of arbitrarily large size would exist. We therefore speak of the "saturation" of nuclear forces, which prevents nuclear matter from collapsing to a density less than  $\rho$  and from flying apart.

 $\mathbf{C}^{\mathsf{c}}$ 

2. Experiments have shown that the forces between two protons, corrected for the effect of Coulomb repulsion, are the same as between two neutrons, i.e., charge symmetry prevails, and probably the forces are also the same between a proton and a neutron (charge independence).

In recent years a number of theoretical physicists, under the leadership of K. Brueckner, have tried to understand the nuclear phenomena by treating the nucleus as a manybody system assuming that nuclear forces can be derived from a two-body potential. The simplified case of infinite

3.24 <sub>y |,38</sub> - 7,2<br>(ლა2) - Fi  $4.23$  $54.26$ 28.29006  $\mathbf{r}$ Mg 24 Mg 26 Mg 27 Mg 25 Mg 23 78.8  $10.1$  $\overline{111}$  $9.5m$  $12s$ 8" 15,157<br>7.84,107,18  $\overline{\mathbf{a}}$  $\sim$  $.27$ 23.9926 24.99375  $\frac{1}{2}$ 25.99080  $\bullet$ **No 23 No 24 Na25 No 22** No 21  $15.0h$ 60s  $2.6y$ 100  $234$ 1139.  $4.0, 2.6 - 3.4$  $54, ...$  $14.1$ 2753136 98.58.38 150  $\frac{28}{2.84}$ 22.9970  $F = 1$  $\mathbf{r}$ **Ne 21 Ne 22** Ne23 Ne24  $40<sub>s</sub>$  $3.4<sub>m</sub>$ **B.9**  $0.26$ 8'1,95, 1-4,3<br>7.470, 88, --<br>E 2.42 l" 4,2, 3,8,<br>, 44, 1,65, 2159635  $F2I$ F 20 II s  $5s$ 14 542  $5.7$ 019 By permission of  $291$ General Electric Co.  $3.2, 4.4$ <br>(970, 1.37)  $79$ 12 m

> nuclear matter is considered first. A few months ago P. S. Signell and R. E. Marshak and independently J. Gammel and R. Thaler showed that the phase shifts derived from nucleon-nucleon scattering experiments up to 150 Mev can be fitted remarkably well by a charge-independent potential including a spin-orbit term of the same sign as in shell theory. This potential has already been introduced in a Brueckner-type theory by de Dominicis, and reasonable answers for the binding energy of infinite nuclear matter have been obtained.

> One would hope to apply similar methods to the more complicated case of the finite nucleus, thereby demonstrating the validity of the shell model for such a system. The next decade may see an understanding of complex nuclei based on interactions derived from nucleon-nucleon scattering experiments.

> While the solution of these very difficult fundamental problems progresses slowly, experimental nuclear physicists are guided in their research by a number of more phenomenological nuclear models, each one of which is limited in its application. Usually these models are not born in a finished form but have to be modified continuously to fit the facts. Sometimes, they even seem to contradict fundamental principles, as when the basic assumption of the shell model that a nucleon may be considered to move in a central potential seemed at first to violate the idea that the mean free path of a nucleon is of the order of its diameter. J. H. D. Jensen, in a talk given in 1956 at the International Congress on Theoretical Physics in Seattle, recounted how Maria Mayer and independently Haxel, Jensen and Suess revived and modified-by the introduction of strong spin-orbit coupling--the early shell-model ideas (which in turn were conceived in analogy to the atomic case) merely as a working hypothesis. A possibility of removing the apparent contradiction was first

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pointed out by Weisskopf, who emphasized the importance of the role of the exclusion principle in simplifying the particle motions in the presence of strong forces. Later quantitative studies of Brueckner, Eden and Bethe have shown that this conjecture is correct and that the shell model can be used to give a useful first approximation to the nucleus up to  $\sim$  10 Mev.

Two other models which have played an important role in recent years are the collective model developed by A. Bohr and B. Mottelson, which is likewise valid at low energies only, and the optical model, which successfully describes nuclear absorption and scattering processes, e.g., the so-called mountain resonances for neutrons and protons.

Let us now consider some of the aspects of these models which are likely to lead to further progress in nuclear research: The shell model will, of course, continue to be useful for the construction of level schemes and in giving the characters (spins and parities) of levels not only of stable nuclei, but also of radioactive nuclei, in providing a means for the classification of beta decays and for the explanation of the occurrence of nuclear isomers, etc. Of more farreaching importance are the possibilities for a quantitative analysis of the energies of nuclear levels. In the near future such an analysis will be restricted to the immediate neighborhood of doubly magic nuclei, e.g., O<sup>16</sup> and Pb<sup>208</sup>.

The successful analysis by Elliott and Flowers, by D. Kurath and by B. French and others not only of the level energies of some light nuclei, but also of the transition rates of a number of beta transitions, and of several electromagnetic transitions-mainly of dipole character-justifies the expectation of further progress in this field.

As an example of a field where much progress can be expected in the near future, and in which the author has been particularly interested, a few words may be said about those even-even nuclei which have vibrational level schemes and which lie between the "magic" nuclei and the rotational region. The vibrational level schemes have been tentatively interpreted on the basis of the Bohr-Mottelson model in the region of weak to moderate coupling. Such a model predicts a triplet, of characters  $0+, 2+, 4+,$  at about twice the energy of the first 2+ state. Since states differing in spin by 4 would escape detection with most of the usual methods of determining level schemes, it is not



#### G. Scharff-Goldhaber

Physics Department, Brookhaven National Laboratory; has worked on nuclear energy levels, neutron and electron diffraction, spontaneous fission, and ferromag $netism.$ 

surprising that only one such close-lying triplet has been found so far, namely, in Cd<sup>114</sup>. The method used here was the analysis of conversion electrons accompanying neutron capture in Cd<sup>113</sup> (H. Motz). A systematic search for triplets by means of suitably chosen experiments should throw light on the question of the nature of the vibrational even-even nuclei and, in turn, of those odd-A nuclei of which they form the core.

As is well known, the collective model has been enormously successful in describing not only the level characters and energy ratios, but also the transition probabilities of electromagnetic transitions, the log  $ft$  values, etc., in strongly deformed nuclei.

Recent efforts to interpret the level scheme of  $\mathbf{F^{_{19}}}$  have led to an interesting discovery: the results of one group of physicists who applied a shell-model analysis agreed surprisingly well with those of another group who applied a collective-model interpretation. Ironically, the theoretical values agreed even more closely among each other than with the experimental values. The reason for the good agreement is by no means obvious and is now being studied by a number of theoreticians. It allows one to conclude, however, that it will be possible to set up a unified model of the nucleus, in which the individual particle motions and the collective motions are self-consistent as in molecules. Important attempts in this direction have been made by Peierls and Yoccoz and by Wheeler and Griffin.

Now a few words about higher-energy nuclear reactions: As was mentioned above, the optical model is successful in describing the energy dependence of the absorption and scattering cross sections of the particles impinging on a nucleus for a considerable energy range. However, a theory has still to be evolved which will give the probabilities and angular correlations for the decomposition of the system target nucleus plus bombarding particle into the various energetically allowed end products. It may be added that the optical model is based on the assumption that the energy spectrum of the incoming particles overlaps many resonance levels in the target nucleus. It is likely that with increasing energy definition this approach will give way to a renewed interest in the fine structure, which is of particular importance in the fission process.

#### We can look forward to definitive progress understanding the bewildering toward variety of phenomena observed in fission

A few thoughts on beta-decay theory: the recent revolution in thought brought about by the discovery that parity conservation and the conservation of charge conjugation do not hold in weak interactions has attracted great interest to this field. Although at present the nature of the interactions for the nucleon-electron-neutrino system is not known, it is very probable that by the end of the coming decade it will be quite well understood. This will be brought about by studies of polarization of the electrons emitted in various types of beta decay, by further efforts to detect double beta decay and by the refinement of present neutrino-detection experiments.

The experimental determination of nuclear properties will increase in accuracy and scope as the equipment and methods grow in diversity and ingenuity.

For example, the steady improvement in resolution and efficiency of spectrometers will facilitate the determination of level energies.

For character assignments to nuclear energy levels there are now a number of methods at our disposal, which we have just begun to exploit. For short-lived states preceded by some previous radiation there is the delayed-coincidence method, which Sunyar has recently developed into a form that permits one to measure lifetimes as low as a few times 10<sup>-11</sup> sec. For electric dipole or quadrupole transitions leading to the ground state the Coulomb-excitation method is able to cover a wide lifetime range. As a final example, the use of molecular-beam and paramagnetic-resonance methods for the spin determination of radioactive nuclei will doubtless increase and serve to check assignments made on the basis of decay-scheme studies. Further, it may be expected that the refinement of the theoretical interpretation of experimental results, e.g., a better theory of stripping, of internal conversion and of the angular distribution of inelastically scattered particles, will make level-character assignments of excited states more definite.

Radioactive nuclei will be found further away from the stability region, owing to the use of heavier bombarding particles and faster detection methods. The study of the binding energies of nuclei of this type may throw further light on the nature of nucleon-nucleon forces. Also, the number of known elements will certainly be increased.

#### Relatively long-lived isotopes may well be found among the far-transuranic nuclides 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  $j$  neutron shell).

New accelerators like variable-energy cyclotrons and tandem Van de Graaffs capable of producing beams of well defined and sufficiently high energies will make it possible to study radiation and particle widths of excited states of light nuclei with accuracy, thus testing the shell-model wave functions, and to explore the level schemes of medium-weight nuclei (50 <  $A$  < 150).

Atomic-beam methods and possibly the study of  $\mu$ -mesic X-rays will yield new data on electric and magnetic moments of higher order. The boldness of some thinkers in this field is indicated by the title of a recent theoretical paper: "Nuclear Hexadecapole Moments." It may even be possible to get a better idea of the charge and current distribution within the proton by means of the 6-Bev electron synchrotron now being constructed at Cambridge, Massachusetts, which will permit an extension of the very successful work carried out at Stanford.

One important goal of nuclear-physics research is the deduction of nuclear forces from meson fields. Among these fields the role of the  $\pi$ -meson field will probably be first understood, but it is clear that any ultimate theory will not be able to ignore the role of the "strange particles" (K mesons and hyperons). In the meantime, the new field of "hypernuclear" physics is likely to develop comsiderably and to help indirectly in the understanding of nuclear phenomena.

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In Nucl. Phys. 81 (1966) 1, Myers and Swiatecki present a mass table based on a postulated shellcorrection expression with  $Z = 126$  as the next magic proton number beyond  $Z = 82$ . However they state that Meldner suggested to them that  $Z = 114$  was another possibility. They also observed that in previously published Nilsson modified-oscillator level diagrams there were large spherical gaps at this proton number (but since they were not labeled their significance was not always recognized).

## **ARKIV FÖR FYSIK**

**UTGIVET AV** 

KUNGL. SVENSKA VETENSKAPSAKADEMIEN

**Band 36 nr 69** 

PROCEEDINGS OF THE LYSEKIL SYMPOSIUM 1966

C. GUSTAFSON, I. L. LAMM, B. NILSSON and S. G. NILSSON

Nuclear deformabilities in the rare-earth and actinide regions with excursions off the stability line and into the super-heavy region



ALMQVIST & WIKSELL **STOCKHOLM** 

1967





On the other hand, for a more complicated r-dependence than the one assumed in eq. (1), one may not be able to conserve the volumes of all equipotential surfaces simultaneously. One may then have to be content with conserving a weighted average of these surfaces.

In the actual calculations it is convenient to make a small change of eq.  $(1)$  by first transforming to the variables  $\xi = x\sqrt{M\omega_1/\hbar}$ ,  $\eta = ...$ , etc., according to ref. [7]. The parameters  $\varepsilon_{20}$  and  $\varepsilon_{40}$  are replaced by  $\varepsilon$  and  $\varepsilon_4$ . We then write the total single-particle Hamiltonian:

616

It was assumed that relatively stable nuclei near the next magic number would be separated from the last stable nuclei at the end of the currently known ones by a sea of instability. Some large-scale calculations happened to include some such nuclei (although they were focused on actinides and the next assumed magic numbers). Some nuclei in the sea of instability were actually calculated to have large negative shell corrections below -5 MeV. But the significance was not noted at the time. (next 4 frames).



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#### ON THE NUCLEAR STRUCTURE AND STABILITY OF HEAVY AND SUPERHEAVY ELEMENTS <sup>†</sup>

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#### Received 14 February 1969

Abstract: Nuclear potential energy surfaces as a function of deformations are calculated on the basis of a modified oscillator model. In particular, quadrupole  $(P_2)$  and hexadecapole  $(P_4)$  deformations are considered. The average behavior of the surface is normalized to that of a liquid drop through the employment of a generalized Strutinsky prescription. In this way a synthesis of the single-particle model and the liquid-drop model is obtained.

Lowest minima in the potential energy surfaces give the ground state masses and distortions. These results compare extremely well with experimental data. Spontaneous fission half-lives are also obtained. The inertial parameters associated with fission barrier penetration are derived empirically as well as by a microscopic model. Shape (fission) isomeric states are also found. Their  $N$  and  $Z$  dependence in the present model are discussed and results tabulated.

The calculations are extended to the predicted superheavy region around  $Z = 114$  and  $N =$ 184. The total overall stability with respect to alpha and beta decay, and spontaneous fission is found to be most favorable in the vicinity of  $Z = 110$  and  $N = 184$ . Detailed diagrams and tables are exhibited.

#### **Introduction**

It was found a long time ago  $1, 2, 3$ ) that simple equilibrium calculations based on the deformable shell model <sup>4</sup>) were able to reproduce the experimental quadrupole moments in the "rare earth" and "actinide" regions. In the Mottelson-Nilsson calculations<sup>1</sup>) single-particle energies are simply added as a function of the quadrupole distortions, and the shape corresponding to the minimum energy is found. The calculations neglected the effects of Coulomb and pairing interactions, which are, however, considered by Bès and Szymański<sup>2</sup>), Szymański<sup>5</sup>), and also by Sobiczewski<sup>6</sup>). On the whole, the results of ref.<sup>1</sup>) are reproduced, indicating that Coulomb and pairing forces at the equilibrium point counteract each other. In these latter

<sup>&</sup>lt;sup>†</sup> Work supported by the U.S. Atomic Energy Commission, the Swedish Council of Atomic Research and the Polish Atomic Energy Commission.

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#### $1.D.2$

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#### **CALCULATED GROUND-STATE PROPERTIES OF HEAVY NUCLEI**

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Received 10 April 1974

Abstract: Ground-state distortions and single-particle corrections are calculated for nuclei with  $Z \ge 68$  and  $N \ge 106$  by use of the macroscopic-microscopic method as developed by Strutinsky. The microscopic part is calculated primarily by use of the folded Yukawa singleparticle potential. Its parameters are redetermined to fit actinide data. The modified oscillator potential is also used in some of the studies. Two methods for calculating the macroscopic energy are investigated. One is the droplet model of Myers and Swiatecki, and the other is a modified liquid-drop model in which the surface-energy term is modified to take into account the finite range of the nuclear force. Single-particle level diagrams for the folded Yukawa potential are also presented. They are plotted as functions of the distortion parameters  $\varepsilon$ ,  $\varepsilon_4$ and  $\varepsilon_6$ . Theoretical and experimental single-particle levels at the ground state for actinide nuclei are also compared.

#### 1. Introduction

Today many calculations of nuclear properties are carried out by use of the macroscopic-microscopic method, which was developed in its present form by Strutinsky  $<sup>1</sup>$ ).</sup> In this method the total potential energy of the nucleus is expressed as the sum of two terms, a macroscopic term and a microscopic term. A recent review of the calculation of fission barriers<sup>2</sup>) contains some results obtained by use of the folded Yukawa single-particle potential to calculate the microscopic corrections and the droplet model of Myers and Swiatecki<sup>3-5</sup>) to calculate the macroscopic part of the energy. A more detailed study of the results obtained for fission barriers is presented in ref. <sup>6</sup>). However, compared to the large distortions involved in fission, much more experimental information is available at ground-state distortions. In this paper we use primarily the folded Yukawa single-particle potential to study single-particle levels, nuclear shapes, and single-particle corrections at ground-state distortions. These quantities depend not only on the single-particle potential but also, to a lesser extent, on the method that is used to calculate the macroscopic energy. We present results obtained by use of the droplet model of Myers and Swiatecki<sup>3-5</sup>) and alternatively by use of a modified liquid-drop model in which the surface-energy term is modified to take into account the finite range of the nuclear force <sup>7</sup>). Details on various aspects of the calculations can be found in refs.<sup>2,7-9</sup>) and in references quoted

<sup>&</sup>lt;sup>†</sup> On leave from the University of Lund, Lund, Sweden.

<sup>&</sup>lt;sup>11</sup> This work was supported by the US Atomic Energy Commission and the Swedish Atomic Research Council.

#### **GROUND-STATE PROPERTIES**



The ground-state single-particle correction is the nuclear ground-state mass relative to the spherical macroscopic energy, which is calculated here by use of the modified-surface-energy model. In this table the ground-state zero-point energy (which is frequently taken equal to 0.5 MeV) is not<br>included. Single-particle levels for  $2\frac{24}{68}$ Ra and  $2\frac{50}{90}$ Cf are used to calculate the potential energy for<br>ne

One then arrives at the following expression for the macroscopic energy in the modified-surface-energy model:

$$
E_{\text{macro}} = -a_v(1 - \kappa_v I^2)A + E(\xi_v) + \frac{4}{3}\pi V_0 R_0^3 + \frac{3}{5} \frac{e^2 Z^2}{r_0 A^{\frac{1}{3}}} \left[ B_C(\xi_v) - \frac{5}{6}\pi^2 \frac{d^2}{r_0^2 A^{\frac{1}{3}}} - \frac{0.7636}{Z^{\frac{1}{3}}} \right].
$$
 (3)

In 1975 Seeger and Howard published the first global mass table with deformations and level structure based on *calculated* microscopic correction based on a general nuclear-structure model and the Strutinsky method. No other mass table in this issue is based on a general theoretical nuclear structure model able to model many correlated nuclear properties.

# These Number Nor Atomic Data Volume 17 Fairleys 5 6 M - June 197 **Nuclear Data Tables**

#### **1975 MASS PREDICTIONS**

S. Maripuu, Special Editor

**TOP** 

EDITOR: Katharine Way MANUSCRIPT EDITOR Lu W. Rose ASSOCIATE EDITORS: E. U. Baranger **B.** Bederson F. Boehm J. W. Cooper A. Dalgarno S. Datz **B.** S. Dzhelepov R. Geballe J. A. Harvey W. C. Lineberger J. B. Marion E. W. McDaniel F. K. McGowan E. Merzbacher M. Mladjenovic G. R. Satchler S. Shafroth

#### Introduction Development of the Semiempirical Droplet Model. W. D. Myers 411 A New Semiempirical Shell Correction to the Droplet Model (Gross Theory of Nuclear Magics). H. v. Groote, E. R. Hilf,  $\overline{418}$ and K. Takahashi A Semienpirical Liquid-Drop Plus Shell-Correction Formula.<br>P. A. Seeger and W. M Howard 428 A Semiempirical Shell Model Formula, S. Liran and N. Zeldes  $.431$  $442$ An Ab-Initio Shell-Model Formula. M. Bauer -Masses of Spherical Nuclei from the Energy-Density Method. 450 M. Beiner, R. J. Lombard, and D. Mas Updated Mass Predictions from the Garvey-Kelson Mass Rela-455 tions. J. Jänecke Ensemble Averaging of Mass Values. E. Comay and I. Kelson  $463$ Masses from Inhomogeneous Partial Difference Equations.<br>J. Jänecke and B. P. Eynon 467 A 1975 Midstream Atomic Mass Evaluation, A. H. Wapstre, 474 and K. Bos........ 476 EXPLANATION OF TABLE ... 477 TABLE. 1975 Mass-Excess Predictions .

### **ACADEMIC PRESS New York and London**

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In 1981 a global mass table based on the folded-Yukawa single-particle model was published. In the summer of 1982 in the LBL cafeteria Peter Armbruster asked Peter Moller: "Do you think the large (negative) shell corrections you obtain in the vicinity of  $\tilde{Z} = 108$  and  $N - 162$  are related to the just discovered new elements at the GSI". The calculated shell corrections were subsequently plotted in color where the large shell corrections for DEFORMED nuclei in the previously assumed "sea of instability" clearly stand out. (next 3 frames)

#### ATOMIC MASSES AND NUCLEAR GROUND-STATE DEFORMATIONS CALCULATED WITH A NEW MACROSCOPIC-MICROSCOPIC MODEL

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We tabulate the atomic masses and nuclear ground-state deformations of 4023 nuclides ranging from <sup>16</sup>O to <sup>279</sup>112, calculated on the basis of a Yukawa-plus-exponential macroscopic model and a folded-Yukawa microscopic model, with new terms included to account for several previously neglected physical effects. With the values of only five constants determined from a least-squares adjustment to ground-state masses, the resulting root-mean-square error in the calculated ground-state masses of 1323 nuclides ranging from <sup>16</sup>O to <sup>259</sup>No for which experimental values are known with experimental errors less than 1 MeV is 0.835 MeV.

\* Alexander von Humboldt Senior U. S. Scientist Awardee

#### TABLE. Calculated Ground-State Electric Multipole Moments and Masses, Compared to Experimental Masses Where Available See p. 171 for Explanation of Table





Fig. 2c. Plot of the ground-state microscopic correction, as calculated in [6] for 4023 nuclei. The fluctuations in the shell correction are larger in the heavier region than in the lighter region.

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The next 5 frames compare calculated  $Q_{\alpha}$ with experimental data. The FRDM data were all published/submitted before the experiments so they represent actual predictions. Many features of the data are reproduced by the FRDM, notice in particular the kink in the vicinity of  $Z = 108$  and  $N = 162$ .











Beginning in 1999 computer tech became sufficiently powerful to make practical to calculate fission potential-energy surfaces versus 5 independent shape coordinates for a total of millions of different shapes. The calculations showed results in agreement with the old assumptions that many actinide systems divide into one large spherical fragment and one smaller deformed. In 2011 Randrup et al (PRL 106, 132503 (2011)) showed that remarkably accurate fission yields could be calculate based on random walks on these potential energy surfaces without introducing any adjustable parameters. Next 4 frames. The random walk method was then used to calculate, or predict, where asymmetric fission would occur, 5th frame.





## Contrasting Fission Potential-Energy Surfaces Hg 
iges>U



The next 4 frames illustrate barrier heights and fission half-lives. The first shows that fission half-lives (shorter than 30d) have mainly been observed for barrier heights above 5 MeV. The next two refer to a Polish Woods-Saxon calculation of barrier heights. The 4th frame are barriers calculated in the folded-Yukawa. Frame 1 and 4 are from PRC 91 (2015) 024310. Frame 4 shows that barriers in the r-process path or decay back from the r-process are so low that fission would occur. The two different models in frame 3 and 4 both indicate that barriers are calculated to be too low to allow nuclei with approximately  $Z > 120$  and  $N > 190$ to exist. It would be desirable to study isotopes with neutron numbers closer to  $N =$ 184 than currently available to test these theoretical predictions.



#### Adiabatic fission barriers in superheavy nuclei

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Using the microscopic-macroscopic model based on the deformed Woods–Saxon single-particle potential and the Yukawa-plus-exponential macroscopic energy, we calculated static fission barriers  $B_f$  for 1305 heavy and superheavy nuclei  $98 \leq Z \leq 126$ , including even-even, odd-even, even-odd and odd-odd systems. For odd and odd-odd nuclei, adiabatic potential-energy surfaces were calculated by a minimization over configurations with one blocked neutron or/and proton on a level from the 10th below to the 10th above the Fermi level. The parameters of the model that have been fixed previously by a fit to masses of even-even heavy nuclei were kept unchanged. A search for saddle points has been performed by the "imaginary water flow" method on a basic five-dimensional deformation grid, including triaxiality. Two auxiliary grids were used for checking the effects of the mass asymmetry and hexadecapole nonaxiality. The ground states (g.s.) were found by energy minimization over configurations and deformations. We find that the nonaxiality significantly changes first and second fission saddle in many nuclei. The effect of the mass asymmetry, known to lower the second, very deformed saddles in actinides, in the heaviest nuclei appears at the less deformed saddles in more than 100 nuclei. It happens for those saddles in which the triaxiality does not play any role, which suggests a decoupling between effects of the mass asymmetry and triaxiality. We studied also the influence of the pairing interaction strength on the staggering of  $B_f$  for odd- and even-particle numbers. Finally, we provide a comparison of our results with other theoretical fission barrier evaluations and with available experimental estimates.

DOI: 10.1103/PhysRevC.95.014303

#### I. INTRODUCTION

Although fission barrier heights  $B_f$  are not directly measurable quantities, i.e., are not quantum observables, they are very useful in estimating nuclear fission rates. As the activation energy  $E_a$  (per mole) in chemistry gives a rate k of a chemical reaction at temperature T via the Arrhenius law  $k = Ae^{-E_a/RT}$ (where  $R$  is the gas constant and  $A$  is the frequency factor) [1,2], the fission barrier gives the fission rate  $\Gamma_f$  of an excited (as they usually are in nuclear reactions) nucleus via:  $\Gamma_f \sim e^{-B_f/kT_{\text{eff}}}$ , where  $T_{\text{eff}}$  is an effective temperature derived from the excitation energy, and  $k$  is the Boltzmann constant. For example, knowing fission barriers of possible fusion products helps to predict a cross section for a production of a given evaporation residue in a heavy-ion reaction: one can figure out whether neutron or alpha emission wins a competition with fission at each stage of the deexcitation of a compound nucleus. Moreover, one can try to understand the experimentally established intriguing growth of the total cross sections around  $Z = 118$ ; for its correlation with  $B_f$ ; see, e.g., Fig. 6 and the related discussion in Ref. [3]. On the other hand, the prediction of the spontaneous or low-energy (i.e., from a weakly excited state) fission rates, governed by the regime of the collective quantum tunneling, requires an additional knowledge of the barrier shape and mass parameters.

A nonobservable status of the fission barrier, again in analogy to that of the activation energy in chemistry, is

A general idea is that, at the excitation energies close to and higher than the barrier but still not inducing sizable dissipative corrections, the adiabatic barrier could be used for calculating fission rates.

Since calculations of potential-energy surfaces (PESs) for odd-A and odd-odd nuclei involve a repetition of calculations for many low-lying quasiparticle states which multiplies the effort (especially in odd-odd systems), systematic studies of their fission barriers are rather scarce. Up to now, they were provided mainly by the Los Alamos microscopic-macroscopic (MM) model and recently by some self-consistent models [5]. The current state of theoretical predictions in fission of even-even nuclei (with  $Z \ge 100$ ) has been discussed recently in Ref. [6].

In the present paper we extend our MM model based on the deformed Woods–Saxon potential, which up to now was applied mainly to even-even nuclei [7], to odd-A and odd-odd

reflected in its possible dependence on a reaction type and/or the excitation energy (effective temperature) range. This leads to some uncertainty in calculations of fission barriers. In particular, it is not clear whether intrinsic configurations should be conserved along the level crossings, which increases  $B_f$ , or the adiabatic state should be followed. This is especially relevant for odd-A and odd-odd nuclei, in which sharp crossings of levels occupied by the odd particle exclude the strictly adiabatic scenario. It is known that, if the projection of the single-particle angular momentum  $\Omega$  on the symmetry axis of a nucleus is conserved, the diabatic effect on the fission barrier can be huge; see, e.g., Ref. [4]. As there is no accepted formula for a barrier correction due to the nonadiabaticity, it is usually ignored, even in odd-N and/or odd-Z nuclei.



FIG. 19. Calculated fission-barrier heights  $B_f$  for superheavy nuclei.





# C O N C L U S I O N S

- All global nuclear-structure models are based on "effective" forces. To expect infinite accuracy with global models is unrealistic.
- Both Wood-Saxon and folded-Yukawa based models give properties of SHE elements to useful accuracy. Both models predicted subsequently observed  $Q_{\alpha}$  values well, in particular the kink near  $Z = 108$  and  $N = 162$ , but show no hope of stability for new elements with  $Z > 120$  and no isotopes with  $N > 190$ .
- Remaining differences between these models and between the models and experiment to a large extent reflect somewhat unavoidable model uncertainties.
- Obviously smaller deviations can be achieved by local adjustments of parameters, but for those of us who strive to improve global model accuracy, this would be a null results.
- Most HFB models have poor (mass) results for known nuclei, therefore their stability predictions in the SHE region are irrelevant. It would be desirable to reduce the number of parameter sets and not use different parameter sets for different purposes, and work to understand how to bring the model results into useful agreement with known data such as nuclear masses.
- For heavy systems it is not the lowest minimum that is the most stable, it is the minimum with the highest fission barrier, a fact overlooked in some calculations.
- Although much used, 2D FISSION potential-energy surfaces obtained in constrained HFB calculations are flawed and do not, and cannot, reflect the properties of the "full" higher-dimensional potential energy function.
- The folded-Yukawa model has been extended to describe many fission properties such as fission (charge, isotopic) yields and neutron-emission versus fragment mass, with encouraging accuracy.

List of publications relevant to Moller presentation 2022-06-15 Three sections: Ground-state properties, beta-decay and astrophysics, fission

#### GROUND-STATE PROPERTIES

- 1. Calculated ground-state properties of heavy nuclei, P. Möller, S. G. Nilsson and J. R. Nix Nucl. Phys. A229 (1974) 292
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