Shape isomers and the double-humped barrier

How four independent puzzles in nuclear physics converge in a single, unifying explanation—some nuclei can exist in two different shapes.

David D. Clark

Efforts to express the total binding energy of the nucleus in terms of the atomic number, the number of neutrons and the shape of the nucleus form an old and continuing search. An accurate expression for this function would help us understand many nuclear phenomena, for example the fission barrier: Under what conditions and in what ways do nuclei fly apart?

Recent evidence that actinide nuclei from thorium (Th, $Z = 90$) to berkelium (Bk, $Z = 97$) possess two equilibrium shapes has inspired a good deal of new work on nuclear binding and suggested some new experimental directions. The discovery of the second shape startled nuclear physicists, who had not expected more than one—the slightly distorted ground-state shape, which is nearly a prolate spheroid with major semiaxis a about 25% greater than minor semiaxis c. The second shape, discovered in several kinds of experiments that we shall discuss, is much more elongated, being roughly a spheroid with a about 80% greater than c.

As we see in the black curve of figure 1 (called the "double-humped fission barrier"), a nucleus in the second shape is metastable. When trapped in the outer minimum, it has an excitation energy 2–3 MeV above the ground state, but its decay is hindered by the inner and outer potential barriers. This new type of metastability is called "shape isomerism" to distinguish it from spin-forbidden isomerism—the slowness of gamma decay between energy levels that differ in angular momentum by several units of $\hbar$.

A qualitatively new phenomenon usually generates interest simply because it is new. In the present case there is an additional reason: The same theoretical models that describe observed effects in the actinides also predict the decay properties, and therefore the possible existence, of long-lived "superheavy" nuclides with atomic number greater than 105. The greater the success in explaining the actinides, the stronger the faith in the predicted parameters for the superheavies.

Evidence for the hypothesis of shape isomerism (or the double hump) comes from four different lines of experimental and theoretical research. One of the fascinating aspects of the new development is how these initially independent endeavors, each encountering its own puzzles, converged upon a single explanation that clarified and united them all.

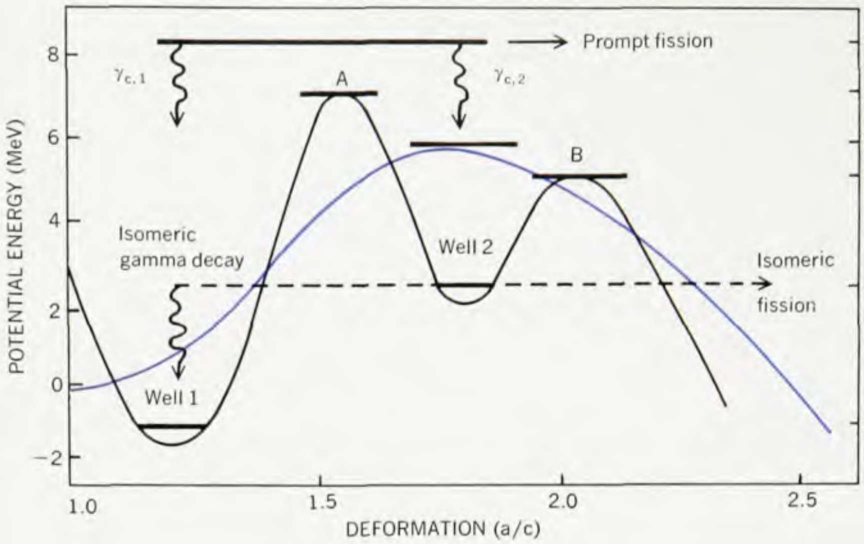
One major category of experimental support is the existence of isomers that decay by spontaneous fission; these are interpreted as nuclei caught in the second minimum that tunnel through the outer barrier, breaking apart into two fragments. Theoretical evidence comes from calculations of the total nuclear binding energy of deformed nuclides when the model includes the effects of single-particle (shell-model) energies; curves of the form of figure 1 result from these calculations. Another line of experimental evidence is the energy dependence of the cross section for fission induced by neutrons or by the (d,p) reaction; two types of resonance structure are observed, and both can be correlated with parameters describing the double-humped barrier. The fourth kind of evidence comes from the interpretation, based on Aage Bohr's "channel" theory of fission, of fission-fragment angular distributions.

A summary of most of the experimental evidence is provided in figure 2, which is a section of the usual nuclide chart. Nuclides in which fissioning isomers have been observed are indicated by a 10^{-n} entry, which gives the order of magnitude of the observed halflife, in seconds. Nuclides in which cross-section resonances have been correlated with a double-humped barrier are shown by a colored nuclide symbol. For some nuclides both types of data exist. The theoretical calculations, which so far have been carried out only for even-Z, even-N nuclides, predict the double-humped barrier for the entire region shown in the figure.

Spontaneous fission

Spontaneous fission from the ground state, that is, tunneling from the ground state through the entire potential barrier, has been known since 1940. It occurs with measurable decay rates in ele-

David Clark is a professor of applied and engineering physics and director of the Ward Laboratory of Nuclear Engineering at Cornell University, Ithaca, New York.



Potential energy of deformation varies with ratio a/c of the major and minor semiaxes of a prolate nucleus. Black curve is the double-humped fission barrier; colored curve is the fission barrier for the traditional liquid-drop model. A nucleus in well 2 with energy E_2 is isomeric (metastable) because of the potential barrier on either side. Decay to the right is by fission, to the left by gamma emission. Energy E_1 is the ground state of well 1; E_2 is the ground state of well 2; E_A, E_B and E_{LDB} are the heights of the inner barrier A, the outer barrier B and the liquid-drop barrier; E_{CN} is the excitation energy of the compound nucleus. Competing de-excitation processes (top) for the compound nucleus are gamma decay into wells 1 and 2 ($\gamma_{c,1}$ and $\gamma_{c,2}$) and prompt fission. Figure 1

ments from uranium on up, the partial halflives decreasing rapidly as Z increases. For example, in uranium isotopes experimental values range from 10^{14} to 10^{17} years, and in californium from 61 days to 10^{9} years. Above californium the partial halflives are shorter still, and spontaneous fission begins to compete with alpha and beta decay in determining the total ground-state halflife. Several systematic correlations of experimental spontaneous-fission decay rates with Z and N have been developed. Extrapolation of these correlations to elements above 104 led people to believe, until only a few years ago, that spontaneous fission in those elements would be so fast that their synthesis would be impossible.

The first discovery of an *isomer* that decays by spontaneous fission was reported in 1962 by S. M. Polikanov and his group[1] at Dubna in the USSR. It was an unanticipated result of a search for new elements formed in heavy-ion bombardment of high-Z elements. Polikanov's group identified the fissioning nuclide as Am^{242}, and the halflife was determined to be 13.5 ± 1.5 millisec, which is at least 10^{19} times shorter lived than predicted by systematics for ground-state spontaneous fission of Am^{242}.

The importance of this anomalous result was immediately recognized, and informed speculations on reasons for such an enormous enhancement of the fission rate appear even in their first paper. According to one hypothesis (which turned out to be partially correct) the decay occurs from an excited isomeric state of Am^{242}. Because the tunneling probability through a potential-barrier curve like the colored one of figure 1 has an extremely steep energy dependence, an excitation energy of only about 2.5 MeV above the ground state corresponds to the observed enhancement factor of 10^{19}. However, a state at this energy would be expected to decay by gamma emission in times much shorter than milliseconds unless some hindrance mechanism were acting.

Evidence for low spin

One such mechanism is spin-forbiddenness, but an unusually high spin, $20\hbar$ or more, would be needed to cause the observed halflife. The puzzle of the anomalous Am^{242} decay therefore deepened when in 1967 G. N. Flerov[2] reported experiments that indicated a low spin value. Flerov and his colleagues measured the ratio of isomer production to the production of the ground state (the "isomeric ratio") as a function of the average angular momentum $\langle l \rangle$ of the compound nucleus. Different reactions as well as different bombarding energies were used to vary $\langle l \rangle$; figure 3 is a plot of their data.

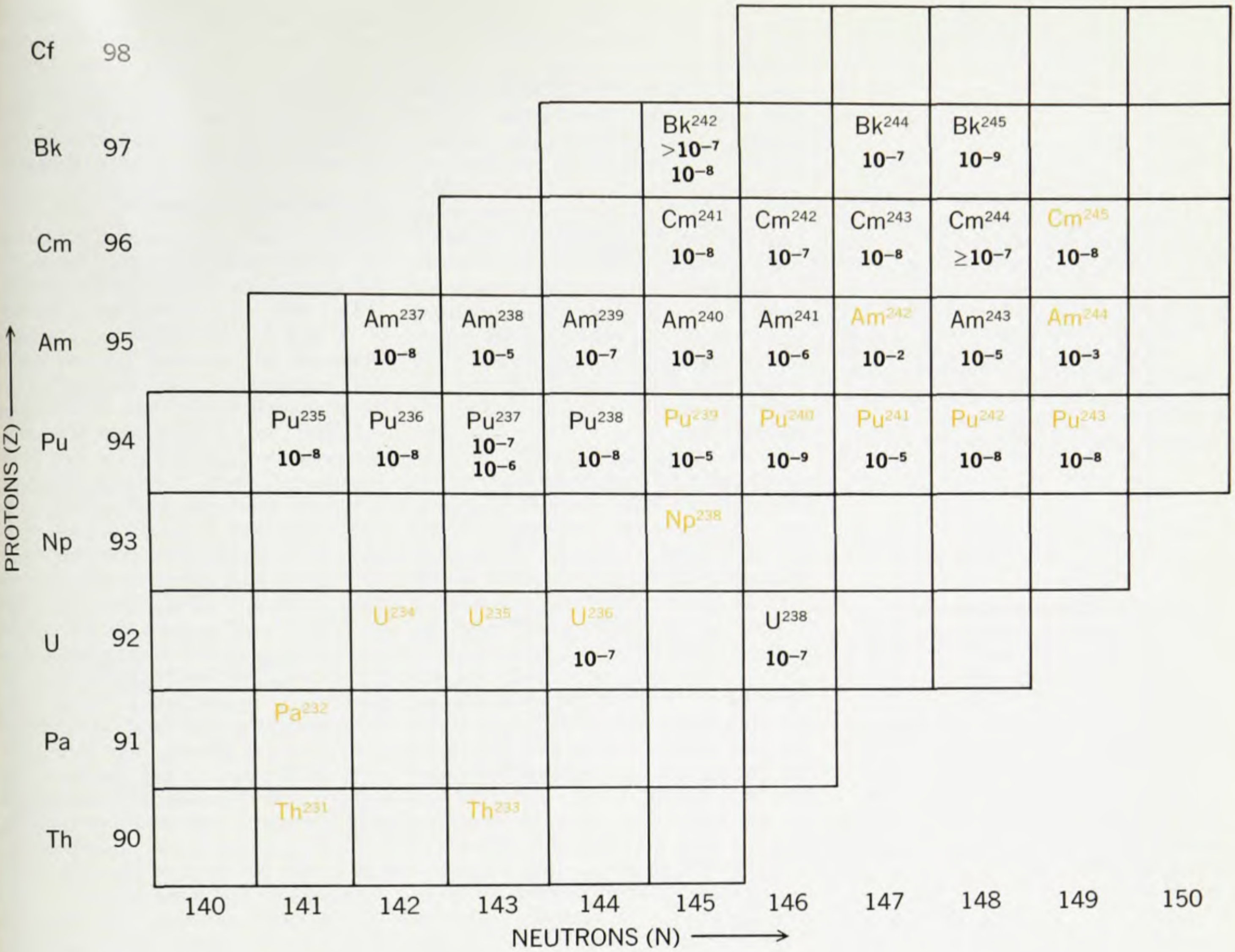
According to the theory of John Huizenga and Robert Vandenbosch,[3] the isomeric ratio for a high-spin isomer should increase monotonically with $\langle l \rangle$ up to values of $\langle l \rangle$ close to the spin of the isomer. Flerov's curve for the well known spin-forbidden Au^{196} isomer with spin and parity 12^- does show the behavior expected for a high-spin state. The curve for Am^{242}, however, does not, so that some new mechanism must account for the exceptional hindrance to gamma decay.

In the meantime, the generality of the phenomenon of fissioning isomers had been suggested by Dubna reports[4,5] of two more confirmed cases, Am^{240} and Am^{244}, both with halflives of about one millisecond. And the indirect estimate of excitation energy derived from the tunneling probability was confirmed in more direct fashion. Sven Bjørnholm and his coworkers[6] at the Niels Bohr Institute in Copenhagen measured the energy threshold for production of the Am^{240} isomer in the (p, 2n) reaction on Pu^{241} to be 3.15 ± 0.25 MeV higher than the threshold for the ground-state reaction, and they interpreted that value as the excitation energy of the isomer. A similar experiment in Bucharest by a joint Russian–Rumanian group[7] investigating formation of the Am^{242} isomer in the (n, 2n) reaction on Am^{243} yielded an excitation energy 2.9 ± 0.4 MeV for Am^{242}. (A recently developed theory by S. Jägare[8] for interpreting threshold measurements requires reduction of these excitation energies somewhat, by about 0.5 MeV.)

All of the experimental methods mentioned so far relied on the impact of energetic beam particles to give the struck nuclei enough recoil energy to carry them completely out of targets as thin as a few micrograms per cm^2. The recoiling metastable nuclei were adsorbed on the surface of a wheel or ribbon moving continuously past the target and were thus carried out of the beam and past a series of fission-sensitive detectors. Various detectors were used, including ionization counters, nuclear emulsions, semiconductor detectors and glass or mica plates. These plates are track detectors, in which the trail of radiation-damaged sites created by the heavily ionizing fission fragments can be enlarged by chemical etching, so that they are visible under a microscope. Halflives were determined from the decrease in the number of fissions as the collecting surface moved away from the target. The usefulness of this technique is limited mechanically to halflives greater than about one millisecond.

Shorter-lived isomers

To search for shorter-lived isomers, different approaches were necessary. In one of these, a thicker target traps most of the recoils and the bombarding beam is pulsed on and off; isomer fission events can then be observed by counters during the intervals between beam pulses. The Copenhagen group[9]



Actinide region of the nuclide chart, summarizing experimental evidence for the double-humped barrier. Halflife entries, in seconds, indicate nuclides for which fissioning isomers have been observed; colored nuclide symbol indicates nuclides for which cross-section resonances have been correlated with the double barrier.
Figure 2

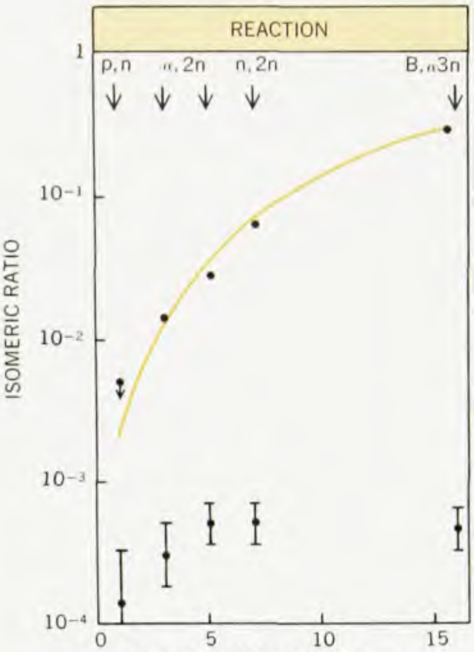
used this method in the discovery of the fourth fissioning isomer, Am^{238}, which has a 60-microsec halflife.

The investigations of the 1962–67 period are lucidly summarized in an excellent review article by Polikanov[5] that appeared in January 1968. Even though two major new developments lay just ahead, the main features of the *experimental* characteristics of the isomers as we now understand them were known even then, and his article is still worth reading. (The prospective reader is warned, however, that three of the "isomers" he lists—Np^{228}, Am^{232} and Am^{234}—are now believed to be nuclides that decay from their ground state by electron capture to a highly excited level of the daughter, which then fissions promptly. A fourth isomer—"Cf^{246}"—has since been reassigned to Cm^{243}.) His discussion of various hypotheses to account for the hindrance of gamma decay makes interesting reading even in the light of what we now believe true.

A major development of 1968 was the general realization that the double-humped barrier predicted by theoretical work, independently in progress for several years, could explain the fissioning isomer phenomenon as well as other puzzling experimental results. In this theory the mechanism hindering gamma decay is simply the inner potential barrier (A in figure 1). Some of the excitement attending this realization is revealed in the *Proceedings*[10] of the international symposium on nuclear structure held at Dubna in July 1968.

The second major development in 1968 was the discovery by Neil Lark and his coworkers at Copenhagen[11] of no less than *ten* new cases of fissioning isomers in uranium, plutonium and americium. The halflives ranged from 1.5 microsec down to about 5 nanosec, much shorter than for the earlier known isomers. The experimental technique that allowed observation of such short halflives is seen in figure 4. Based on a method described by Yu. P. Gangrsky, it used large-area plastic foils as fission-track detectors viewing a stream of collimated recoils from a thin target. The geometric arrangement shielded the plastic foils from fission fragments that emerged directly from the target due to prompt fissions occurring there. The plastic detectors thus recorded only delayed fissions that took place while the nuclei were in flight, and halflives were calculated from the estimated recoil velocity and the density of fission tracks as a function of distance from the target.

These results were important both in establishing the general occurrence of fissioning isomers in the heavy elements and in encouraging other searches for additional isomers. The present total of 27 nuclides exhibiting fission isomerism, shown in figure 2, includes contributions from groups at Seattle, Heidelberg, Los Alamos and Argonne, as well as Copenhagen and Dubna.[5,10,12–17] A variety of nuclear reactions and bombarding particles and energies have been used, but the isomeric



A measure of isomer production, isomeric ratio, varies with angular momentum $\langle l \rangle$ of the compound nucleus. The ordinary high-spin isomer Au^{196} shows a continually increasing ratio (color), whereas Am^{242} levels off at low $\langle l \rangle$, indicating low spin in the isomeric state. This low spin is evidence that spin forbiddenness could not be hindering gamma decay, so that some new explanation was needed for the halflife of Am^{242}. The data come from the four indicated nuclear reactions.
Figure 3

ratio is always small, of the order 10^{-4}–10^{-6}. Another ratio, that of delayed (isomeric) fissions to prompt fissions, is usually of the same order, causing the experimenter considerable difficulty. The smallness of these ratios can be explained in terms of competition between prompt fission, capture into the first well and capture into the second well. Several other important measured, or still to be measured, characteristics of the isomers will be discussed later.

Existing nuclear models

Theory, as we have noted, was also undergoing new developments in the search for an accurate expression for the total nuclear binding energy, or "potential-energy surface" as a function of N, Z and shape. But before we look at models giving the double-humped barrier, we should recall some earlier theories of the nucleus. A nucleus is a system of $A = N + Z$ fermions that acts like a drop of a very cold (that is, degenerate) Fermi liquid. The properties of this liquid, neglecting surface and Coulomb effects, have been derived by the Brueckner–Bethe–Goldstone theory of infinite nuclear matter from the two-body nucleon–nucleon forces determined from scattering experiments. Good agreement with empirically deduced values is achieved for fundamental parameters such as the liquid density. Calculation of the potential-energy surface for real finite nuclei in terms of nuclear matter theory would, however, be extremely complicated; instead, the usual approach has been to seek semiempirical expressions. Nuclear matter theory does, nevertheless, give us some insight into the empirical nuclear properties on which such expressions are based.

One basic property of nuclear matter, empirically deduced many years ago, is "saturation"—the principal phenomenon underlying the liquid-drop model. Saturation describes the observation that inside all nuclei (except the smallest), all nucleons have the same binding energy (15 MeV per nucleon) and the same number density (0.2 nucleon per fm^{3}). Nuclear-matter theory shows that these characteristics arise from the specifics of the strength and shape of the nucleon–nucleon potential—primarily the combination of repulsive core and the short-range, strong, attractive potential—together with the consequences of the exclusion principle. Furthermore, the theory shows that the interparticle spacing and potential are such that the "state" of nuclear matter in nuclei is liquid rather than solid. In common with other saturating systems of a finite number of particles, nuclei also display surface effects: Empirically, the density drops to zero in a layer that is thin compared to the radius of medium and heavy nuclides, and we find a surface "tension" (1.3 MeV/fm^2).

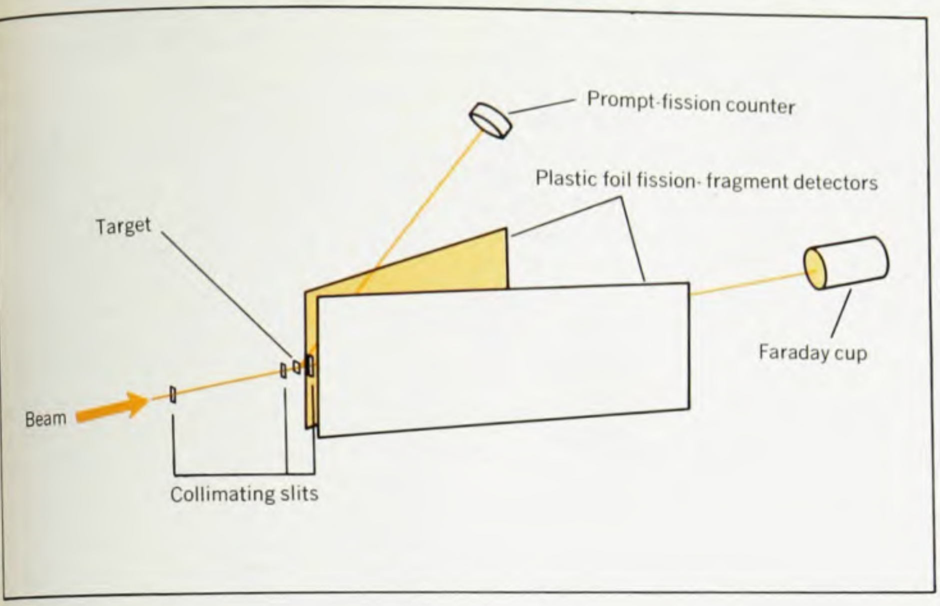
A second key empirical fact about nuclei is that the mean free path of a low-energy nucleon moving through nuclear matter is relatively long compared to a nuclear radius, a necessary condition for the independent-particle model, in which individual nucleons move within the nucleus in "orbits" with well defined quantum numbers. Models that describe the shell structure in both spherical and deformed nuclei are based on the independent-particle model. A long mean free path might be (and for many years was) thought impossible in a dense medium bound by strong forces, but nuclear matter theory explains it, as it does saturation, on the basis of the nucleon–nucleon force and the exclusion principle. The latter plays a particularly obvious role: It forbids a moving nucleon to interact with any other nucleon in any way that would put either particle into an already occupied state. Because the nucleus is a very cold Fermi system, most states below the Fermi level are indeed occupied, so that only a nucleon with initial energy above the Fermi level can scatter from another nucleon. L. C. Gomes, John Walecka and Victor Weisskopf[18] have given a particularly clear discussion of how nuclear matter theory can explain both saturation and the long mean free path.

Semiempirical treatments

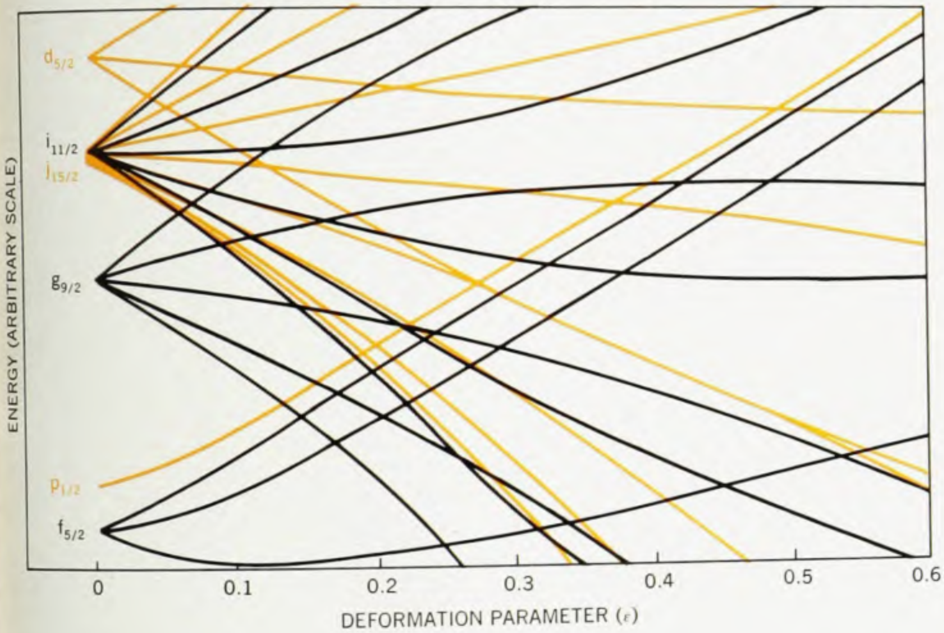
The earliest semiempirical expression for the potential-energy surface was the von Weiszäcker–Bethe–Bacher formula (1936) for nuclear masses as a function of N and Z. Dependence on nuclear shape was not included, because nuclei were assumed to be spherical before the discoveries of fission and of deformed nuclides. In its usual form the semiempirical mass formula has five binding-energy terms: volume, surface, Coulomb, symmetry and pairing. The first two are due to the saturation property, and the Coulomb term expresses the antibinding effect of the electrostatic repulsion of the protons. The symmetry and pairing terms, which have no analogues in a macroscopic liquid drop, express the tendency of nuclei to be more tightly bound when $N = Z$ (symmetry) and when either N or Z or both are even numbers (pairing). When the five adjustable parameters of the formula are determined experimentally, the resulting function follows very closely the smooth overall trend with N and Z of the more than 1200 experimental mass values.[19]

There are, however, two ways in which it fails in detail. In the immediate neighborhood of the magic numbers of the spherical-shell model (N, Z = 2, 8, 20, 50, 82, 126), experimental values show a small but distinct systematic dip below the smooth curve of the formula. The maximum deviation is a few millimass units; in terms of energy, the deviation is a few MeV out of a total binding energy of the order of 8A MeV, or about 0.5%. And, between magic numbers, the experimental masses lie slightly above the curve; the deviations, which are small, show systematic trends related to the occurrence of nonspherical ground-state shapes (such as in the rare earths and the actinides) that have been understood since the work of Aage Bohr and Ben Mottelson in the 1950's. Both types of disagreement can be associated with the occurrence of shell structure in nuclei.

The same semiempirical formula, emphasizing as it does the liquid-drop analogy, can also treat the fission barrier. Here N and Z are constant and the shape is changed. If we consider the energy needed to deform a nucleus from an equilibrium spherical shape, at constant volume, only the surface- and Coulomb-energy terms have any effect. They have opposing tendencies under deformation: The potential energy is increased as the surface area increases,



Short-lived fissioning isomers (1.5 microsec–5 nanosec halflives) were detected by Neil Lark and his coworkers at Copenhagen with a special target and detector arrangement (left). The plastic foils are geometrically placed to record only delayed fissions that occur while the nuclei are in flight, not the prompt fissions that occur directly in the target. Halflives are calculated from the estimated recoil velocity and the variation of track density in the foils with distance from the target.
Figure 4



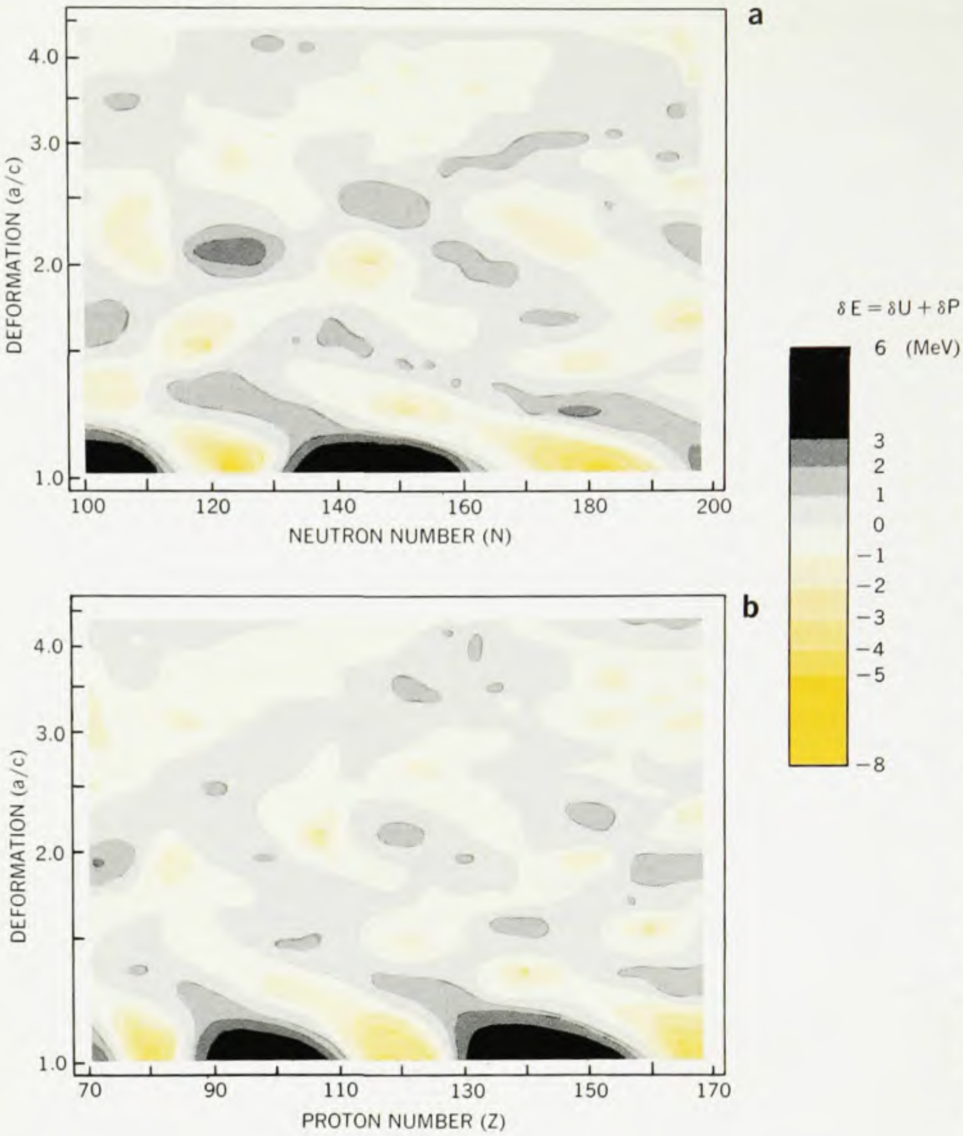
Neutron energy levels in a Nilsson potential. Deformation parameter ϵ, which is zero for a spherical nucleus, expresses the degree of prolateness of the spheroidal nucleus. Spherical-model levels, designated by l_j symbols, are $(2j + 1)$-fold degenerate but split under deformation into $(2j + 1)/2$ doubly degenerate levels of different m_j. For simplicity, levels originating from higher and lower spherical-model states are not shown. The number of energy levels per unit energy interval is seen to vary with occupation number N and with deformation.
Figure 5

whereas the Coulomb term leads to a decrease, because the protons are moved farther apart on the average. The combined effect is a function of the form indicated by the colored curve in figure 1. The specific functional form of course depends on the detailed shape of the deformation as well as on the parameters and functional forms of the surface and Coulomb energy terms. The double-humped barrier deviates from the smooth liquid-drop curve because of shell structure effects that are, as we shall see shortly, basically the same as those that produce the mass deviations described above. In this sense, the wiggles in the mass curve and those in the fission barrier are closely related.

Independent particles

At this point we should note the salient features of the shell models for spherical and deformed nuclides. These independent-particle models treat each nucleon as moving independently in a potential well formed from the average attraction to all the other nucleons. In a complete microscopic treatment, this "nucleon–nucleus" potential would be a self-consistent one derived from nuclear-matter theory, but the calculational complexities are prohibitive. Instead, we choose a suitable semiphenomenological potential well (giving up the self-consistent property) and calculate two sets of energy states in the well, one for the neutrons and one for the protons. The ground state of a nuclide with given N and Z is then represented by filling in the states, starting with the lowest, in accordance with the exclusion principle until (at the Fermi level) the number of neutrons and protons is exhausted. The primary application of shell models is to properties of the ground level and the low-lying excited levels that involve only nucleons in the last few filled states. Properties that are successfully correlated and predicted include ground- and excited-state spins and parities; level ordering and spacings, and the transition rates and selection rules for gamma and beta decay.

The first successful shell model, that of Maria Goeppert-Mayer and J. Hans D. Jensen, deals with spherical nuclei. For deformed nuclei, the most famous shell model is the Nilsson model,[20] which assumes an anisotropic harmonic-oscillator potential plus a spin-orbit term and an additional term, proportional to l^2, that truncates the potential appropriately. In the original form of the model, the deformation was limited to quadrupole (P_2) shapes, and the requirement of constant nuclear density was met approximately by demanding that the volume enclosed by any given equipotential surface be conserved as a function of deformation. In Nilsson's first paper, Coulomb and pairing interactions were neglected, but later they were included by other authors. The adjustable parameters of the model



Contour maps show the variation of the sum δU (the shell-correction term) plus δP (the pairing energy correction) with nucleon number and nuclear deformation. According to the "Strutinsky prescription," the correction sum is added to the smooth liquid-drop model energy (E_{LD}) to give a potential-energy surface that fits the observed mass variation with N and Z quite well. Note that the sum is strongly negative for both neutrons (a) and protons (b) in the actinide region at $a/c \approx 2$, in agreement with observations, and again in the superheavy region ($Z \approx 115$, $N \approx 183$) at a/c about one.
Figure 6

are fitted to experimental data. Typical results[21] are shown in figure 5 for neutron orbitals in the region around $N = 126$. Here the abscissa is a deformation parameter that expresses the degree of prolateness of the assumed spheroidal shape, so that the curves show the single-particle energies as a function of deformation. At zero deformation ($\epsilon = 0$) the level ordering and spacings are the same as for the standard spherical-shell model.

In addition to satisfactorily treating single-particle properties, the Nilsson model reproduces quite closely the experimentally determined ground-state shapes of deformed nuclei. In contrast to this success, however, when the deformation is increased away from equilibrium to large values, the predicted total binding energy does not vary in a manner that comes even close to reproducing a fission barrier. This failure of the otherwise remarkably successful Nilsson model has been attributed to an unsatisfactory mathematical description of large deformations and to the more basic and intractable reason that the assumed nucleon–nucleus potential is not the correct self-consistent one.[19,22] One consequence is that the Nilsson model fails to reproduce saturation.

Combining the models

Our discussion began with the problem of finding an accurate formula for the potential-energy surface. As we have just seen, neither the liquid-drop model alone nor the shell model alone is adequate. The liquid-drop semiempirical mass formula, however, *does* closely represent the smooth overall trend of masses, and the shell effects are in a sense small fluctuations about this trend. Thus an obvious approach is to start with the drop formula and improve it by adding shell and deformation "correction" terms with additional adjustable parameters. Although most proposed modifications have treated only ground-state masses, a few workers in the mid 1960's, among them V. M. Strutinsky in the USSR, William Myers and Wladyslaw Swiatecki[19] in Berkeley, and Nilsson's group[21] at Lund (Sweden), were also concerned with the potential-energy surface at large deformations, such as in fission.

During the period 1965–68 Strutinsky,[23] working first in Moscow and later in Copenhagen, achieved the first successful quantitative amalgamation of the liquid-drop and shell (Nilsson) models for large deformations; the set of calculational steps that he developed has become famous as the "Strutinsky prescription." He first makes the common assumption that the smooth trend of the energy surface is correctly given by the liquid-drop model or, more specifically, by the volume, surface and symmetry terms of the semiempirical formula plus a readily calculated Coulomb term. This contribution to the energy is called E_{LD}. To E_{LD} he adds a shell-correction term δU and a pairing term δP. The sum gives an energy surface that fits the measured mass variation with N and Z quite well. It also predicts the double-humped barrier and shows that the second minimum is due to δU, because the variation of δP with deformation is smooth.

Strutinsky's main contribution was his prescription for calculating δU. To understand the method, we note that in shell-structure theory the existence of regions of increased total binding energy (shells) has often been ascribed to the degeneracy of single-particle levels due to spherical symmetry. But from a more general viewpoint introduced by Strutinsky (and also in different form by Myers and Swiatecki), shell structure is a reflection of irregular variations in the "density" of single-particle energy levels near the Fermi level. First, we note that the number of energy levels per unit energy interval (the level density) fluctuates considerably with energy. In figure 5, for example, we see that at zero deformation there are

$2j + 1 = 10$ levels at exactly the same energy for the $j = 9/2^+$ state, and large gaps with no levels at other energies. Next, we define a smooth "average" level-density distribution function by a weighted average $\langle g \rangle (E)$ of the single-particle energies over a suitably chosen energy interval centered on E; Strutinsky has shown that with proper choices of weights and interval, $\langle g \rangle(E)$ is a smooth function of E. To connect shell structure and level density we then note that if the density in the neighborhood of the Fermi level E_F is lower than $\langle g \rangle(E_F)$, the total nuclear binding energy is larger than the liquid-drop value, and the converse is true if the level density is higher than $\langle g \rangle(E_F)$. Thus a closed shell is indicated when the level density is high just below and low just above the Fermi level; inspection of figure 5 shows this to be true, for instance, for a spherical nucleus with 126 neutrons, the number needed to fill completely all levels through the $f_{5/2}$ and $p_{1/2}$ levels. From this generalized definition of shell structure it easily follows that the long-familiar wiggles in mass around the drop-model mass curve can have a closely related counterpart in wiggles in deformation energy around the smooth drop-model fission barrier. In other words, the double-humped barrier can occur because the level density at the Fermi level for a given nucleus varies with *deformation* as the nucleus traverses the thicket of energy levels in the Nilsson diagram of figure 5.

Strutinsky defines the shell-correction term δU as the *difference* between the sum of the energies of the occupied shell-model states and a weighted sum (integral) of those energies, where the weighting function is the smooth level-density function $\langle g \rangle(E)$. The fundamental assumption of the entire method is that $\langle g \rangle(E)$ calculated from the shell model is the same as the single-particle level density that is appropriate to the liquid drop in the region of the Fermi level. In effect, Strutinsky provides a way to "renormalize" the average trend of the (erroneous) single-particle energy sum to the smooth, correct liquid-drop trend.

Strutinsky's first paper on his method appeared in 1966; the work was concurrent with but independent of the experiments on fissioning isomers. But in 1967 he noted that the second minimum in his calculated fission barriers could be the explanation of the isomers. Strutinsky and his colleagues[24] have continued to develop and apply his method, as have Nilsson's group[13] and others.[25] To date, several extensive sets of calculations of the parameters of the double-humped barrier have been published for $Z = 90$ to $Z > 100$. The Woods-Saxon potential, as well as the Nilsson potential, has been used, and deformations other than a simple P_2 distortion have been included. Figure 6 shows, in the form of contour maps, one set of $\delta U + \delta P$, due to Hans Christian Pauli (see reference 12). In all the calculations, one important trend is that for the lighter actinides, the inner hump is lower than the outer, and for the heavier actinides the situation is reversed. The overall theoretical results show that despite the somewhat *ad hoc* character of the prescription, the agreement with experiment is remarkably good, more than merely qualitative.

Cross-section resonances

In the mid-1960's another type of experiment that was being independently pursued in several European laboratories led to unexpected or unexplainable results that were later clarified by the double-hump model. These were high-resolution measurements of heavy-element fission cross sections for neutrons in the eV–keV range and, separately, in the range 0.2–2 MeV.

In both types of experiments the target nuclides were ones for which the compound-nucleus excitation energy produced by the addition of a low-energy neutron would be below the top of the fission barrier; that is, $E_{CN} < E_{LDB}$ in figure 1. Any observed fission would therefore have to occur by tunneling through the barrier, so the cross section should be extremely small at low energies and display a steep monotonic increase with neutron energy.

A typical result[26] for the eV–keV range is seen in figure 7a. The sub-barrier resonances have both a fine structure and an intermediate structure. The fine-structure level density corresponds to the known or expected density of compound-nucleus excited levels at an excitation energy of about 5 MeV, which is the binding energy of the added neutron in the compound nucleus. The intermediate structure is, however, a real puzzle until we invoke the concept of the second well. Consider a nucleus trapped in the second minimum but possessing an excitation energy above the "ground state" E_2 of the second well. The average level density in the second well is less than that in the first well at the same total excitation energy, because the second well is a few MeV shallower (or, alternatively, because some of the total excitation energy is taken up in the deformation, leaving a smaller amount to be shared as ordinary excitation among other modes). The intermediate-structure level density is therefore interpreted to be the level density in well 2; the sets of levels in the two wells are weakly coupled by tunneling through the inner barrier.

This explanation is even semiquantitatively acceptable: If we assume the statistical distribution function for level density to have the same form in the two wells, and also assume a reasonable shape for the second well, we can estimate its depth. The result is that the bottom of the second well is 2 to 3 MeV above that of the first well, in agreement with estimates of E_2 from isomer-excitation functions.

At higher neutron energies, for which the experimental energy resolution is a few tens of keV, the typical resonance structure observed[27] is seen in figure 7b. In pre-Strutinsky days this type of resonance was ascribed to the opening of an inelastic scattering channel at the resonance energy, resulting in a decreased flow in the fission channel. But this explanation is quantitatively unsatisfactory, as pointed out by J. E. Lynn[28] in the final chapter of his book on resonance neutron theory, written before the double hump was known; he had to leave the problem unresolved.

But with the double-hump hypothesis, the resonance could be interpreted as a vibrational resonance in the second well, or, in different words, as a resonance in the tunneling probability for a system penetrating a double barrier. By a nice coincidence of timing, Lynn was able to add the new interpretation to his book at the galley-proof stage, so that the chapter makes particularly fascinating reading. The argument for the tunneling resonance interpretation was clinched in later experiments by Jørgen Pedersen and B. D. Kuzminov,[29] who observed similar resonances in (d,p)-induced fission. Here inelastic channels could not possibly be responsible, because the excitation energy was below the neutron binding energy.

Angular distribution in fission

Our final category of experimental evidence for the double-hump theory is the change in angular distribution of fission fragments with change in bombarding energy and target nuclide. The interpretation of these results is essentially based upon Aage Bohr's channel theory of fission. Consider a nucleus with just enough excitation energy to pass over the top of the fission barrier; at this stage of the fission process, nearly all of the energy is taken up in deformation, and only one or two states of intrinsic excitation are energetically possible for the nucleus. These states or channels have specific angular momenta and parities, which prescribe angular distributions different from the isotropic (or fore-and-aft symmetric) distributions expected from a statistical average over many open channels. The angular distribution should therefore change character quite noticeably with bombarding energy as the number of open channels changes. However, in the case of the double hump, *and if* the inner hump is higher than the outer one, an angular orientation imposed in passing through a *single* channel at the inner hump can be lost in passing over the second well and second hump, where many channels would be open. Because the relative height of the two humps changes monotonically with A, as we noted in the discussion of theory, there should be a corresponding change in the angular distribution results—a prediction that is in rough agreement with the data. (See reference 12 for a summary of these results.)

Directions for research

The experimental and theoretical data we now have strongly support the hypothesis of the double-humped barrier as well as the introduction of a new type of metastability, shape isomerism, and no satisfactory alternative explanation has been advanced. Many questions, however, must be answered before the explanation can be considered proven in detail.

The properties of the isomeric state are only imperfectly known, even in the most-studied examples. For more stringent tests of the theory, the spin, parity, energy, halflife and decay modes of the isomeric level should be determined by the best possible methods for at least a few of the isomers. The only decay mode seen to date is fission, largely because a fission event is rather easily distinguished from most types of background events. But along with this advantage comes the disadvantage that it is well-nigh impossible to determine from fission fragments what the energy, spin and parity of the isomeric level might be. (Experiments by B. H. Erkkila and Robert Leachman,[30] and by Robert Ferguson and his colleagues[31] have shown that the mass and energy distributions of fragments from isomeric fission are indistinguishable from those in ordinary fission; this result is also interesting itself for what it says about the fission process.)

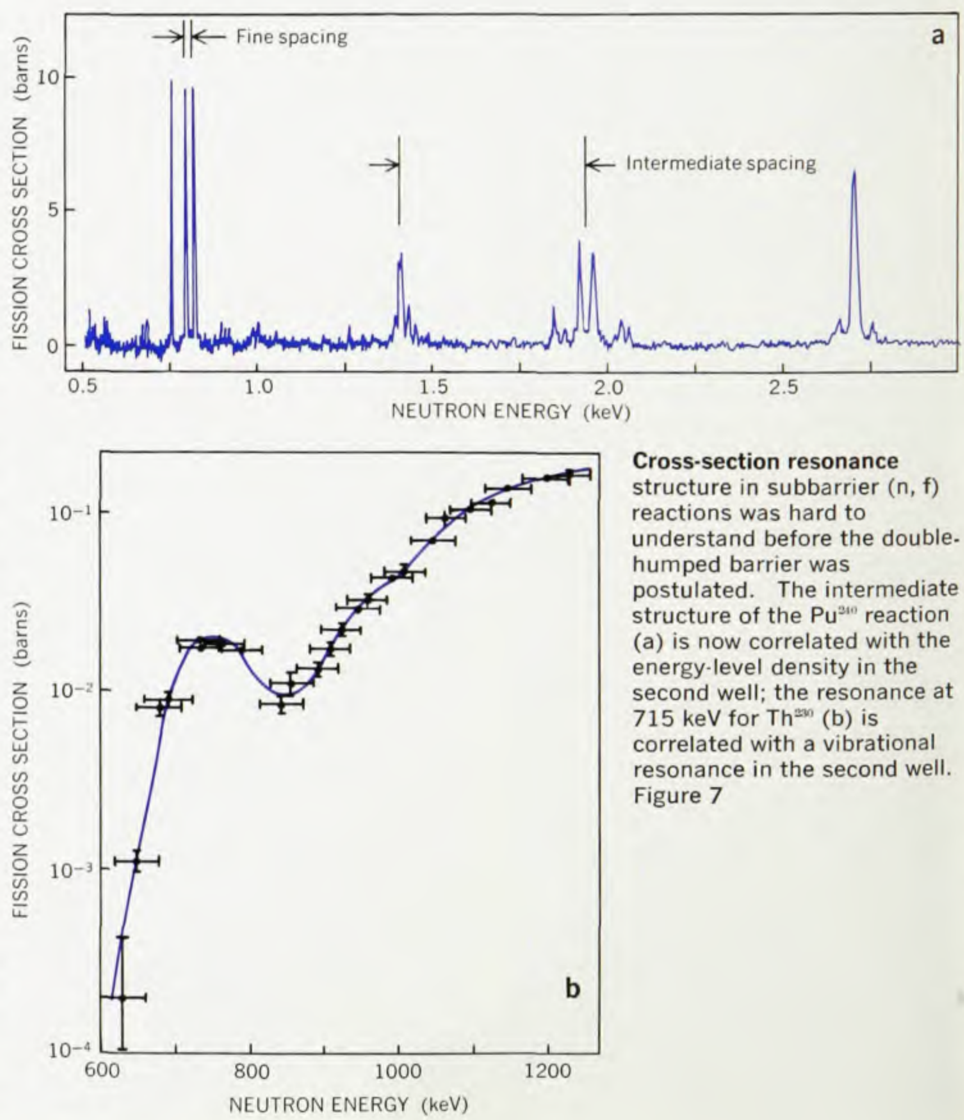
The properties of the isomeric level could be determined by well tested nuclear spectroscopic techniques *if* one could observe the gamma-ray cascade associated with tunneling "back" into the first well (see figure 1). That this decay mode—the "gamma branch"—exists is a virtual certainty if the double-hump hypothesis is correct, but the branching ratio may be very unfavorable in all the known isomers. Several intensive efforts (including one by Hans-Fritz Brinckmann, Pedersen and myself) to find the gamma branch have, because of low yield and high backgrounds, resulted only in setting an upper limit of about ten times the rate of the fission branch. If the theory is correct, however, in the lighter actinides such as thorium the penetrability of the (lower) inner barrier is greater than that of the outer barrier, and gamma branching may be heavily favored over fission. This effect probably accounts for the failure of attempts to find fissioning isomers in isotopes of thorium, protoactinium, uranium and neptunium, for which cross-section resonances imply a double-humped barrier (see figure 2). Gamma-decaying shape isomers should be sought in that region.

Observation of both fission and gamma decay in the *same* nuclide would yield very valuable information on the relative parameters of the two barriers; present theoretical estimates of the branching ratio are very crude. Another useful decay mode would be alpha emission, but theory indicates it is very unlikely. In sum, measurements on the gamma branch are probably the most reliable and straightforward way of establishing the spin, parity and energy of an isomer.

The existence of two fissioning isomers in the same nuclide has been confirmed in at least one case, Pu^{237}. This situation is probably due to two isomeric states in the second well, the "ground" state at E_2 and an excited state whose gamma decay within the second well is hindered by ordinary spin forbiddenness.[32] The phenomenon is worth further investigation.

The probability of formation of fissioning isomers has been rather extensively studied in a variety of charged-particle reactions and to a lesser extent in neutron and photon reactions. One example is a recent lengthy investigation by Harold Britt and his coworkers at Los Alamos,[17] who use energetic deuteron and alpha beams to deduce E_A, E_B and E_2 for the barriers in Pu, Am and Cm isotopes. The analysis provides another, but still indirect, argument for a low spin for the isomeric level. Similar experiments using neutrons at low energies would be especially valuable.

A phenomeon that has only been touched on is the cascade of "capture radiation" (γ_{c2} in figure 1) emitted when the compound nucleus decays into the isomeric level. If these radiations could be identified and measured, we could fix the energy, spin and parity of the isomer. We would also determine some of the excited levels (such as rotational bands) built upon the "ground" state of the second well, and from them we might find parameters of the second well, including the moment of inertia. The moment of inertia would provide a



Cross-section resonance structure in subbarrier (n, f) reactions was hard to understand before the double-humped barrier was postulated. The intermediate structure of the Pu^{240} reaction (a) is now correlated with the energy-level density in the second well; the resonance at 715 keV for Th^{230} (b) is correlated with a vibrational resonance in the second well. Figure 7 unique check on theoretical predictions of the deformation at the second minimum. Such experiments are, however, very difficult because of low yield and high background.

Because theory predicts a double-hump barrier near $Z = 85$, $N = 118$, as well as in the actinide region, experimental searches for fission isomerism there have been undertaken. A recent experiment at Berkeley[33] shows negative results as far as fissioning isomers are concerned but does not rule out gamma-decaying shape isomers.

On the theoretical front, several interesting developments are in progress. Our discussion so far has been limited to the calculations of the potential-energy surface. Fission, however, is a dynamic process, and inertial effects must be taken into account. After all, the entire mass of the nucleus is involved. Thus in treating spontaneous fission, the tunneling probability is affected by an inertial parameter B, which is akin to the alpha mass in alpha-tunneling calculations. Estimates of B and its dependence upon shape have been used by Nilsson[13] to predict spontaneous-fission halflives of both actinide and superheavy elements, with barrier parameters found by the Strutinsky method. The predictions are quite sensitive to the value of B; the present 30% uncertainty in B corresponds to a factor of 10^6 in the halflife! Nevertheless, the predictions of an island of long-lived superheavies around $Z = 114$, $N = 184$ are sufficiently intriguing that superheavies are being sought both in nature and with accelerators, but with no positive proof as yet of their existence.

Regarding the potential-energy surface, there are some investigations of the theoretical foundations of the Strutinsky method, by Wing-fai Lin[34] and by William Bassichis and colleagues. But most of the effort is directed toward application of the method to more realistic potentials for the single-particle states and to more sophisticated parametrization of the shape, which must be described at all stages from the sphere at zero deformation through the cigar, the dumbbell and finally the separated fragments. We do not know the true shapes but must seek to describe them with a manageable number of parameters. (One basis for hope that only a few parameters are needed for an adequate description of the admittedly complex shape has been expressed by Pauli,[14] who noted that an analogous problem in Hollywood has been solved by the use of only three numbers.)

One approach is a two-center shell model proposed by P. Holzer, Ulrich Mosel, and Walter Greiner. (See references in 25; David O. Maharry and J. P. Davidson have proposed an interesting variation on this model.) In their model one calculates the single-particle states in a system of two Nilsson-type anisotropic harmonic oscillators whose centers coincide at zero deformation and undergo increasing separation as deformation proceeds. This model emphasizes the correct asymptotic condition for two separated fragments. It has not yet been possible, however, to impose a satisfactory volume-conservation condition representing saturation in this model without the Strutinsky prescription.[25]

Other groups working on the shape parametrization question, but more directly within the Strutinsky method, include Peter Möller and Nilsson at Lund, James Nix and coworkers at Los Alamos, Jens Damgaard and coworkers in Copenhagen and Pauli and his coworkers at Basel.[35] The most interesting direction in all these efforts is the inclusion of deformations that are reflection-asymmetric about the equatorial plane of the nucleus. All of the earlier calculations have been for reflection-symmetric shapes, which lead only to fission into two equal fragments. The potential-energy surfaces obtained by all these groups indicate that asymmetric deformations are energetically favored and that we may therefore be on the verge of explaining at last the famous unsolved problem of the asymmetric mass division in fission—although the full dynamical treatment remains to be carried out.

* * *

The author is grateful to the Guggenheim Foundation, the Niels Bohr Institute and in particular to Sven Bjørnholm for the opportunity afforded him to learn about shape isomerism during his 1968–69 sabbatic leave spent at the Institute in Copenhagen.

References

1. S. M. Polikanov, V. A. Druin, V. A. Karnaukhov, V. L. Mikheev, A. A. Pleve, N. K. Skobelev, V. G. Subbotin, G. M. Ter-Akop'yan, V. A. Fomichev, Sov. Phys.—JETP **15**, 1016 (1962).
2. G. N. Flerov, Yu. P. Gangrsky, B. N. Markov, A. A. Pleve, S. M. Polikanov, H. Jungclaussen, Sov. J. Nucl. Phys. **6**, 12 (1968).
3. J. R. Huizenga, R. Vandenbosch, Phys. Rev. **120**, 1305 (1960).
4. Yu. V. Lobanov, V. I. Kuznetsov, V. P. Perelygin, S. M. Polikanov, Yu. Ts. Oganesyan, G. N. Flerov, Sov. J. Nucl. Phys. **1**, 67 (1965).
5. S. M. Polikanov, Sov. Phys.-Usp. **11**, 22 (1968).
6. S. Bjørnholm, J. Borggreen, L. Westgaard, V. A. Karnaukhov, Nucl. Phys. **A95**, 513 (1967).
7. G. N. Flerov, A. A. Pleve, S. M. Polikanov, S. P. Tretyakova, N. Martalogu, D. Poenaru, M. Sezon, I. Vilcov, N. Vilcov, Nucl. Phys. **A97**, 444 (1967).
8. S. Jägare, Phys. Lett. **32B**, 571 (1970).
9. J. Borggreen, Yu. P. Gangrsky, G. Sletten, S. Bjørnholm, Phys. Lett. **25B**, 402 (1967).
10. *Proceedings* of the International Symposium on Nuclear Structure, Dubna, 1968 (IAEA, Vienna, 1968).
11. N. L. Lark, G. Sletten, J. Pedersen, S. Bjørnholm, Nucl. Phys. **A139**, 481 (1969).
12. S. Bjørnholm, V. M. Strutinsky, Nucl. Phys. **A136**, (1969).
13. S. G. Nilsson, C. F. Tsang, A. Sobiczewski, Z. Szymanski, S. Wycech, C. Gustafson, I. Lamm, P. Möller, B. Nilsson, Nucl. Phys. **A131**, 1 (1969).
14. *Proceedings* of the Second IAEA Symposium on Physics and Chemistry of Fission, Vienna, 1969 (IAEA, Vienna, 1969).
15. S. M. Polikanov, G. Sletten, Nucl. Phys. **A151**, 656 (1970).
16. R. Repnow, V. Metag, P. von Brentano, Z. Physik **243**, 418 (1971).
17. H. C. Britt, S. C. Burnett, B. H. Erkkila, J. E. Lynn, W. E. Stein, Phys. Rev. C **4**, 1444 (1971); Phys. Lett. **31B**, 523 (1970).
18. L. C. Gomes, J. D. Walecka, V. F. Weisskopf, Ann. Phys. (N.Y.) **3**, 241 (1958).
19. W. D. Myers, W. J. Swiatecki, Nucl. Phys. **81**, 1 (1966).
20. S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **29**, No. 16 (1955).
21. C. Gustafson, I. Lamm, B. Nilsson, S. G. Nilsson, Arkiv. Fysik. **36**, 613 (1967).
22. W. H. Bassichis, L. Wilets, Phys. Rev. Lett. **22**, 799 (1969).
23. V. M. Strutinsky, Sov. J. Nucl. Phys. **3**, 449 (1966); Nucl. Phys. **A95**, 420 (1967); Nucl. Phys. **A122**, 1 (1968).
24. M. Brack, J. Damgaard, H. C. Pauli, A. S. Jensen, V. M. Strutinsky, C. Y. Wong, Rev. Mod. Phys. (to be published).
25. U. Mosel, H. W. Schmitt, Nucl. Phys. **A165**, 73 (1971).
26. E. Migneco, J. P. Theobald, Nucl. Phys. **A112**, 603 (1968).
27. P. E. Vorotnikov, S. M. Dubrovina, V. A. Shigin, G. A. Otroschenko, Sov. J. Nucl. Phys. **5**, 210 (1967); L. G. Earwaker, G. D. James, ref. 14, page 911.
28. J. E. Lynn, *The Theory of Neutron Resonance Reactions*, Clarendon Press, Oxford (1968), chapter 8, sections 11 and 12.
29. J. Pedersen, B. D. Kuzminov, Phys. Lett. **29B**, 176 (1969).
30. B. H. Erkkila, R. B. Leachman, Nucl. Phys. **A108**, 689 (1968).
31. R. L. Ferguson, F. Plasil, G. D. Alam, H. W. Schmitt, Phys. Lett. **31B**, 526 (1970).
32. P. A. Russo, R. Vandenbosch, M. Mehta, J. R. Tesmer, K. L. Wolf, Phys. Rev. **C3**, 1595 (1971).
33. S. Bjørnholm, J. Borggreen, E. K. Hyde, Nucl. Phys. **A156**, 561 (1970).
34. W. Lin, Phys. Rev. **C2**, 871 (1970).
35. H. C. Pauli, T. Ledergerber, M. Brack, Phys. Lett. **34B**, 264 (1971); J. R. Nix, M. Bolsterli, E. O. Fiset, J. Norton, reference 14, page 183. □