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Low-Energy Nuclear Fission and Our Understanding of the Nucleus*

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Abstract

The interactions between experimental discoveries in low-energy nuclear fission and our theoretical understanding of the structure of the nucleus are reviewed. The history of this synergistic relationship begins with the discovery of fission, the development of the liquid-drop model and the experimental evidence for magic numbers, continues through the development of the shell model, the experimental discovery of shape isomerism, the double-humped fission barrier resulting from theoretical calculations of shell corrections to the liquid-drop model, the spontaneous fission half-life disaster, the discovery of symmetric mass division in spontaneous fission and theoretical treatments based on different paths to scission. It concludes with a brief review of our current experimental and theoretical understanding of low-energy fission and the prospects for future developments.

I. INTRODUCTION

In the present paper we review how experimental studies of low-energy and spontaneous fission have affected our theoretical understanding of the nucleus. From the very beginning of the discovery of nuclear fission, the experimental discoveries have outdistanced theoretical predictions. The idea that a heavy nucleus could fission when bombarded with neutrons was so unexpected that in 1934 Fermi[1] believed that he was producing transuranium isotopes upon bombardment of uranium with neutrons. Even at the end of 1938, Hahn and Strassmann were fearful of reporting the results of their painstaking radiochemical experiments showing that they had isolated barium isotopes from the products of neutron irradiation of uranium.[2] In November 1938, Hahn communicated these results to Lise Meitner, a former leader of the team, saying he was certain of the chemistry but not of the physics! She encouraged him saying she knew of no reason why it was impossible. During December, 1938 she and her nephew, Otto Frisch, discussed the surprising results and came up with the idea of division of the uranium nucleus into two large fragments, a process which they called fission. On this basis, they also pointed out that if barium (Z = 56) were one of the products, the complementary one must be krypton (Z = 36).

Hahn and Strassmann subsequently published their results in the January 6, 1939 issue of *Naturwissenschaften*[2] but with the disclaimer that "We, as chemists, based on the briefly described experiments, should rename the above-mentioned scheme and replace Ra, Ac, Th with the symbols Ba, La, Ce. As nuclear chemists, being in some respects close to physics, we have not yet been able to take this leap which contradicts all previous experiences in nuclear physics. It could be that a series of strange coincidences could have mimicked our results." Shortly thereafter, Hahn and Strassmann confirmed the presence of krypton in their experiments.[3] Meanwhile, Meitner and Frisch had submitted an article[4] to *Nature* on January 16, 1939, entitled "Disintegration of Uranium by Neutrons: a New Type of Nuclear Reaction" in which they explained the main features of low-energy fission based on the "present ideas

about the behavior of heavy nuclei" of Bohr[5] in which "the particles in a heavy nucleus would be expected to move in a collective way which has some resemblance to a liquid drop. If the movement is made sufficiently violent by adding energy, such a drop may divide itself into two smaller drops."

Meitner and Frisch estimated the total kinetic energy (TKE) released in this new "fission" reaction to be about 200 MeV, close to the subsequently measured value. They further pointed out that the surface tension of the nucleus decreases with increasing nuclear charge and estimated that it may become zero for nuclei with atomic numbers on the order of 100, thus presaging our current understanding[6] that it is in the region of the heavy fermium isotopes that the liquid-drop barrier to fission disappears and that it is the shell structure of these heavier nuclei that stabilizes them against fission. Shortly after the discovery of neutron-induced fission, Flerov and Petrzhak[7] reported the discovery of spontaneous fission (SF) in uranium, a process which had been predicted by Bohr and Wheeler.[8]

II. THE LIQUID-DROP MODEL TO NUCLEAR SHELLS

A. The Liquid-Drop Model

The interpretation of the Hahn-Strassmann results[2,3] by Meitner and Frisch[4] on the basis of the liquid-drop model gave a remarkably clear picture of the fission process. Subsequently, in their seminal 1939 paper,[8] Bohr and Wheeler gave a more detailed picture of fission and were able to fit most of the experimental data known at that time into a rather satisfactory description of the mechanism of nuclear fission based on a comparison of the properties of the nucleus to those of a liquid drop. This included estimation of the energy release in fission for different charge and mass splits, neutron emission at scission, prompt and delayed-neutron emission from the fission fragments, the critical nuclear deformation required for fission, fission thresholds, the variation of the fission cross section with neutron energy and the fissioning species, the problem

of the statistical distribution of the charge and mass of the fission fragments, as well as fission induced by protons and deuterons. They also predicted the probabilities for fission, neutron emission, and radiative capture as a function of the nuclear excitation energy.

However, the success of the liquid-drop model[8] in predicting the general features of the fission process was tempered somewhat in following years by its inability to provide an explanation for many of the experimentally observed features of low-energy fission. These included the half-life systematics for SF (see Figure 1) and the predominantly asymmetric mass division observed for thermal-neutron-induced and spontaneous fission.

It was shown in von Gunten's[9] 1969 review that all the measured mass-yield distributions for low-energy and spontaneous fission were highly asymmetric and it was commonly believed that this would be the case for all low-energy fission. This asymmetric mass division was widely attributed to "magic numbers," *i. e.*, nuclear shells in the fragments. Magic numbers had been explained much earlier by Mayer and others[10–12] in terms of single-particle orbits and the independent-particle shell model; this theoretical explanation was developed to explain experimental observations of extra stability for nuclei near the magic numbers relative to that predicted from "smooth" liquid-drop macroscopic properties for nuclei with the magic number of neutrons and/or protons. The experimental data indicated that in low energy and spontaneous fission, mass splits were favored in which one of the primary fragments was near the region of the (Z=50, N=82) magic numbers, and the maximum cumulative yield for the heavy fragment was near mass 134 with the average mass being nearly constant at 140. The position of this average mass of the heavy fragment was found[13] to remain relatively constant for fissioning nuclides from mass 230 to 256 while the position of the light fragment moves from around mass 90 to higher masses as the mass of the fissioning system is increased.

Kelson[14] related the asymmetric mass-yield distributions observed for spontaneous

fission to the addition to the overall nuclear shape of effects due to the intrinsic nuclear structure and the nuclear spin-orbit coupling. Using a prolate deformation representing a major-to-minor axis ratio of 1.4 and a positive quadrupole moment, he was able to fit the basic features of the mass distribution from SF of ^{252}Cf.

B. Neutron Resonances

The reactions of low-energy neutrons with heavy elements has aided the understanding of the fission process since the earliest days of the study of fission. Indeed, fission was discovered[2,4] in neutron irradiations of uranium. However, neutron-induced fission has also proven to be a valuable probe of the structure of the nucleus. The ability to measure neutron velocities accurately (using, for example, time-of-flight techniques) can lead to very precise measurements of reaction and fission cross sections as a function of the incident neutron energy. Additionally, since the neutron experiences no Coulomb repulsion and the neutron-binding energy varies between even and odd nucleons, it is possible to produce compound nuclei with excitation energies below the fission barrier.

Since nuclei can be formed at very low energies where the nuclear structure is still dominated by levels whose widths are smaller than the average spacing between them, it is possible to observe the effects of these levels on the reaction probability. Indeed, early studies of neutron-induced reactions revealed sharp resonances in the reaction cross sections as a function of neutron energy. From scattering measurements, it was possible to assign spins and parities to these resonance states. In addition, some resonance states were observed to be fission resonances as well, *i. e.*, there were sharp peaks in the neutron-induced fission cross sections at those energies as well. From these data, it is possible to determine the fission width (Γ_f) of the resonances, which is related to the fission probability P_f by the relationship

$$P_f = \frac{\Gamma_f}{2\pi\rho}, \tag{1}$$

where ρ is the nuclear level density. For example, Farrell[15] studied the fission resonances of ^{239}Pu and was able to investigate the fission probability of ^{240}Pu as a function of energy, spin, and parity.

It was also suggested by A. Bohr[16] that fission properties such as mass distribution and total kinetic energy release may depend upon the spin and parity of the fissioning state. Cowan *et al.*[17] report variations in mass distributions as a function of the initial state, but searches for effects on the kinetic energy have been negative.[18] These and other aspects of neutron-induced fission are reviewed in depth in the book by Vandenbosch and Huizenga.[19]

III. SHELL MODEL TO SYMMETRIC FISSION

A. Shape Isomerism

In 1962, Polikanov *et al.*[20,21] observed a very short-lived ($\sim$ 0.02-s) fission activity in bombardments of ^{238}U with both ^{16}O and ^{22}Ne. The half-life of this activity was anomalously short and inconsistent with the half-life systematics known at that time (see Figure 1), even if it were an unknown isotope of fermium. Furthermore, this short-lived activity was not observed in an irradiation of ^{232}Th with ^{22}Ne, indicating that the activity was not likely to be a compound nucleus residue since this reaction yields the same compound nucleus as the reaction of oxygen with uranium. Polikanov hence assigned this activity to $Z < 100$, probably resulting from some type of transfer reaction.

The low Z assigned, however, exacerbated the problem of the short fission half-life. According to the systematics known at the time,[22] the lighter elements should have fission partial half-lives on the order of hundreds of days to thousands of years. To reconcile their results with the contemporary understanding of the nucleus, Polikanov *et al.* suggested that the fission might be occurring from a long-lived isomeric state, which would see a smaller effective fission barrier and hence fission more rapidly than

the ground state. Such a state could be analogous to high-spin α-decaying nuclei which had been observed to have a high hindrance for α decay.[23,24]

Within a few years, many anomalously short-lived fission activities had been discovered and the hypothesis that these were isomeric states in known nuclei had been confirmed by various techniques (See, for example, the paper by Polikanov and Sletten[32]). However, there remained the question of what sort of isomers these were. Neither α nor γ decay from the fission isomer had been observed, so information on the spin and energy of the isomer was unavailable.

In 1967, Bjørnholm *et al.* measured the excitation energy of the isomeric state $^{240}\mathrm{Am}^f$ ($t_{1/2} = 0.6$ ms) produced by the $(p, 2n)$ reaction.[45] They observed a definite threshold for the production of the fission isomer at a bombarding energy of about 11 MeV while the ground state production threshold was at about 7.5 MeV. This corresponded to an excitation energy for the fission isomer of 3.15 ± 0.25 MeV. At this high an excitation energy, though, it was impossible to reconcile the reaction cross-section data with the very large spin required for the isomer to be γ stable. Adding to the dilemma, Flerov *et al.* studied the production of $^{242}\mathrm{Am}^f$ as a function of angular momentum introduced by the incoming projectile and found that the isomer to ground state production ratio was almost independent of the angular momentum introduced.[29] From these results, Flerov *et al.* concluded that the fission isomer must have a low spin.

The combination of these two results eliminated the possibility that these fission isomers were "regular" spin isomers as postulated by Polikanov. At an energy of about 3 MeV and with low spin, γ transitions would depopulate a "normal" isomeric state much too quickly to allow isomer lifetimes as long as the ones observed. Clearly, this was a new type of isomer, with unusual γ-decay and fission characteristics.

This problem was brilliantly resolved by Strutinsky[26,27] in 1968. Strutinsky proposed a shell-correction method to the liquid-drop model in which the shell effects did not disappear as the nucleus deformed. The existence of these deformed shells

explained the deformed ground-state of the actinide nuclei, and explained the fission isomer phenomenon by showing a second minimum in the potential energy surface at higher deformation due to the addition of shell effects. This is illustrated schematically in Figure 2.

The double-humped fission barrier finally reconciled the theory of the nucleus with the measurements of Flerov *et al.*[29] and Bjørnholm *et al.*[45] Since the fission isomer could exist in the second minimum, one would expect γ decay to be severely hindered since it would now require a collective rearrangement of the entire nucleus for γ deexcitation of the isomer, rather than the simpler rotational-vibrational rearrangement for transitions within the same well.

Additional support for Strutinsky's theory was provided in 1969 by Gangrskiĭ *et al.*,[30] who found that the double-humped barrier approach correctly explained their experimental results on the $^{241}\mathrm{Am}(n,\gamma)^{242}\mathrm{Am}^f$ reaction. Lark *et al.* measured excitation energies for a large number of fission isomers in uranium, neptunium, plutonium and americium, and found reasonable agreement with the theory.[31] Polikanov and Sletten expanded the number of observed fission isomers from the uranium isotopes to the curium isotopes[32] in 1970.

Definitive proof of the existence of a deformed second minimum was provided in 1972 by Specht and coworkers.[33] They measured conversion electrons from the 4-ns $^{240}\mathrm{Pu}^f$ isomer which they attributed to three E2 transitions within the isomer's ground-state rotational band. This band was assigned as $K^\pi = 0^+$. Given that the energy for a transition between an initial spin state J_i and a final spin state J_f in the same rotational bandhead is (to first order)

$$E_{J_i \to J_f} = \frac{\hbar^2}{2I}[J_i(J_i+1) - J_f(J_f+1)], \tag{2}$$

where the rotational parameter is $\frac{\hbar^2}{2I}$ and I is the nuclear moment of inertia, the rotational parameter for $^{240}\mathrm{Pu}^f$ was found to be 3.331 ± 0.008 keV. By contrast, the rotational constant for the 0^+ ground-state rotational band of $^{240}\mathrm{Pu}$ is 7.16 keV. This

result indicated that the moment of inertia, I, of the fission isomer was about a factor of two larger than that of the ground state. Hence the fission isomer was indeed much more deformed than the ground state, proving the hypothesis of shape isomerism.

To date, fission isomers have provided fertile ground for investigations of the properties of the fission barrier. Notable achievements include the measurement of the γ-deexcitation branch of $^{238}U^f$, the measurements of level schemes within the second minimum, and the generalization of Specht's measurement of the rotational constant to several other shape isomers. These and other recent advances in shape isomerism have been reviewed by Poenaru, Ivaşcu, and Mazilu.[34] Fission isomers are currently known in isotopes of uranium,[25,35] neptunium,[36] plutonium,[31,37–42] americium,[31,32,43–45] curium,[38,41,43,46,47] and berkelium.[38,41,43]

B. Superheavy Nuclei

Myers and Świątecki published a semi-empirical theory of nuclear masses and deformations in 1966[48] which treated the nucleus as a liquid drop with simple shell corrections. A surprising result of this work was that the model predicted that superheavy nuclei such as $^{310}126$ would be highly stabilized against spontaneous fission by shell effects. For example, the "pure" liquid-drop model predicts that $^{310}126$ would have essentially no fission barrier and a resulting SF lifetime of about 10^{-22} seconds. Assuming that this nucleus is the next doubly-magic one after ^{208}Pb, inclusion of the shell effects would raise the predicted fission barrier by about 9 MeV. A fission barrier this high would mean that this and other superheavy nuclei could be sufficiently long-lived to study by conventional techniques following accelerator irradiations. Indeed, the superheavy elements (or SHE's) might even be stable on a geological time-scale. Other calculations[49,50] indicated that the "island of superheavies" might extend as low as Z = 114.

This, of course, led to an intense experimental effort to synthesize SHE's in accel-

erator experiments or to locate them in nature. Attempts to observe superheavies in terrestrial samples generally proceeded on the assumption that the SHE's behaved similarly to the lighter homolog element(s), *e. g.*, element 118 was expected to behave similarly to radon. This assumption, however, was not necessarily valid and a great deal of effort was directed at predicting the chemical[51,52] and geochemical[53,54] behavior of the SHE's from first principles.

Searches were also performed for superheavy elements in extraterrestrial as well as terrestrial samples. Lunar rock samples, returned by the Apollo landings, were analyzed by both radiochemical means[55] and neutron-multiplicity counting[56] with negative results. Since superheavy elements were expected to be formed in supernovae,[64] they might be detectable in cosmic rays. A number of searches using nuclear emulsions[65] and nuclear emulsions with plastic track detectors[66] were performed, and weak evidence for cosmic rays with atomic numbers of 96 and ~105 was obtained. However, analysis[67] of olivine crystals in meteorites (a long-term *in situ* natural deep-space track detector) revealed no tracks attributable to superheavy elements with a limit of detection two orders of magnitude lower than the nuclear emulsion work.[65,66] Similar analyses of marine sediments agreed[56,68,69] with the results of the meteorite studies.

Accelerator experiments designed to produce and characterize SHE's began almost immediately after the publication of the Myers and Świątecki mass formula. Primary interest in these experiments were directed towards complete fusion reactions using actinide targets and projectiles such as argon,[70] krypton[71] and zinc.[72] All of these experiments gave negative results, either due to the extremely low probability of forming the compound nucleus, the low survivability of the compound nucleus with respect to prompt fission, or both.

In 1972, Bolsterli *et al.*[73] and Fiset and Nix[74] calculated the fission barriers of superheavy nuclei using a macroscopic-microscopic calculation (wherein the smooth or liquid-drop-like aspects of the nucleus are separable from the quantum shell effects)

based on realistic diffuse-surface, single-particle potentials. Their results indicated that the island of superheavy nuclei was of somewhat different shape and location than that predicted by Myers and Świątecki, with the nucleus that was most stable relative to fission occurring at $^{298}114$, in agreement with the calculations of Nilsson *et al.*[50] The island extended to perhaps $^{308}122$ before SF half-lives became precipitously short. Equally important, they predicted that nuclei with $Z < 108$ and $N > 190$ would have essentially no fission barrier. This would terminate the r-process production of superheavy elements before the island of superheavies could be reached. These predictions were confirmed theoretically by Brack *et al.* nearly simultaneously.[75]

Such calculations implied that superheavy residues of supernovae would not survive, and hence one would not expect to find such material in terrestrial nor extraterrestrial matter. However, the gentler slope of the island of stability on the proton-rich side was highly encouraging to attempts to synthesize SHE's in accelerator-based experiments. Even if one could not land on the peak of the island, nuclei "on the beach" might live long enough to be observed.

Hence, a flurry of experiments were undertaken to produce and characterize SHE's. Compound nucleus reactions[76–80] such as $^{48}Ca + ^{248}Cm$ were employed, as were large mass-transfer reactions such as $^{238}U + ^{238}U$.[81,82] Elaborate detection systems such as the **S**mall **A**ngle **S**eparator **SY**stem (SASSY)[83] and the **S**eparator for **H**eavy **I**on **P**roducts (SHIP)[84] have been developed to allow the identification of a single superheavy atom. Painstaking chemical separations have been performed on irradiated samples to search for superheavies down to the picobarn production cross-section regime. All of these searches have proven negative for superheavies to date, although the instruments and techniques developed for this purpose have permitted the discovery of new elements up to $Z = 109$. Experimental searches for SHE's have been recently reviewed by Seaborg,[85,86] Herrmann,[87] and Armbruster.[88,89] Despite the discouraging results of the past twelve years, efforts at producing SHE's are ongoing. One of the most ambitious proposals for SHE production is the **L**arge **E**insteinium

Accelerator Program (LEAP),[90] an American consortium which seeks to increase the United States' annual production of ^{254}Es tenfold (to about 40 μg) in order to use it as a target in SHE experiments.

C. Symmetric Fission in Heavy Fermium Isotopes

Greatly enhanced yields for symmetric mass division were first reported in 1970 for the spontaneous fission of 100-d ^{257}Fm.[91] This was confirmed in 1971[92] and a broadly symmetric mass distribution was measured[93] for thermal neutron-induced fission of ^{257}Fm. It was suggested that this increased yield of symmetric mass splits was related to the fact that fermium with Z=100 could divide symmetrically into two fragments, both having the magic number of 50 protons. These results challenged the accepted view that all low-energy fission gave asymmetric mass splits and caused a renaissance of interest in spontaneous fission properties. Over the next few years, solid-state coincidence techniques facilitated the measurement of many kinetic-energy and mass-yield distributions for shorter-lived, difficult to produce SF activities.

By 1979, mass-yield and kinetic-energy distributions had been measured for the SF of ^{246}Fm, ^{248}Fm, and ^{254}Fm through ^{259}Fm and from neutron-induced fission of ^{255}Fm and ^{257}Fm, *i. e.*, ^{256}Fm* and ^{259}Fm*. The surprising results showed narrowly symmetric mass distributions for ^{258}Fm and ^{259}Fm as shown in Figure 3. These symmetric mass distributions were accompanied by very high average total kinetic energies ($\overline{\mathrm{TKE}}$) which approached the total energy available for fission. As shown in Figure 4 they did not fit on the linear correlations of $\overline{\mathrm{TKE}}$ vs. $Z^2/A^{1/3}$. It was postulated[97,98] that these very high $\overline{\mathrm{TKE}}$'s resulted from the larger Coulomb repulsion due to the nearly spherical fragments which are stabilized by the approach of the fragments to the doubly magic (Z=50, N=82) ^{132}Sn configuration.

The SF properties of ^{256}Fm and ^{258}Fm were also compared with those of the fission of the respective excited nuclei resulting from n-capture. The mass distribution for

the fission of ^{256}Fm* showed increased yields for symmetric mass splits and a slightly higher $\overline{\text{TKE}}$ for symmetric mass splits than for SF of ^{256}Fm. In the case of ^{258}Fm*, the effect was just the reverse, the mass distribution was broadened and was less sharply symmetric. The $\overline{\text{TKE}}$ was considerably lower than for the SF of ^{258}Fm. These apparently conflicting results were again explained on the basis of the fragment shell effects. In the case of ^{256}Fm and lighter actinides, it was postulated that these fragment shell effects tended to stabilize asymmetric mass division while in ^{258}Fm they stabilized symmetric mass division and the extra excitation energy diminished the shell effects. Thus the mass distribution were more symmetric in one case and more asymmetric in the other.

Theoretical attempts to explain these abrupt changes in low-energy fission properties were based on two general approaches—one in which mass division is determined primarily by the deformation energy surface of the fissioning nucleus at the saddle point calculated according to the method of Strutinsky,[26,99–103] and the other based on the shells in the fragments, in which the mass division is correlated[104–106] with the properties of the fragments. Considerable debate ensued as to whether the experimental observations were a consequence of the disappearance to the second barrier to fission in the potential energy surface of the fissioning nucleus, resulting in symmetric mass division, or whether the fragment shell effects were the controlling factor.

D. Symmetric and Asymmetric Fission of Pre-Thorium Nuclei

Shortly after the discovery of symmetric mass distributions in the spontaneous fission of ^{257}Fm, the fission properties of the light actinides were examined for additional information in the attempt to understand the transition between asymmetric fission and symmetric fission. Early measurements of the mass yields of charged-particle induced fission of bismuth[107] and radium[108] had indicated that the mass-yield distribution in the bismuth system (with about 20 MeV of excitation energy) was symmetric

while the mass-yield distribution of radium displayed three peaks. As experimental data had shown by the early 1970's, fission was broadly symmetric up to around lead or bismuth, triple-humped in radium, asymmetric from thorium to fermium, and becoming symmetric again in the heavy fermium isotopes. Pashkevich,[109,110] building on the two-fission mode hypothesis of Turkevich and Niday,[111] suggested that the observed mass distributions in radium were the result of two separate fission paths, *i. e.*, two modes of fission, and was able to obtain qualitative theoretical agreement with the observed mass-yield distribution of radium.

An asymmetric fission mode had been reported in ^{210}Po in a radiochemical study of the fission products of proton-induced fission of bismuth in 1961 by Sugihara *et al.*,[112] so it was conceivable that the triple-humped mass distribution of radium was the end of a broad transition region from symmetric fission to asymmetric fission. However, a subsequent measurement[113] using silicon-surface-barrier detectors to detect coincident fission fragments from proton-induced fission of bismuth failed to confirm the asymmetric component reported by Sugihara *et al.*

In 1982 a very small (0.4%) asymmetric fission branch was reported[114] in ^{213}At. This discovery was shortly followed by measurement[115] of the excitation energy dependence of the probability of the asymmetric fission mode in ^{213}At, where it was found that the asymmetric mode was suppressed with increasing excitation energy. Asymmetric fission was subsequently observed in ^{208,210,212}Po, again with asymmetric fission branches on the order of a few tenths of a percent.[116] In 1985, Itkis *et al.* found that the asymmetric fission mode finally disappeared in ^{201}Tl and proposed that the lower limit for this fission mode was about (Z=80, A=200).[117]

Experimental studies of the asymmetric to symmetric transition region in the pre-actinide nuclei has recently been reviewed by Itkis *et al.*[118] The insights gained from these investigations have essentially confirmed the two-path mechanism of fission in this area as proposed by Pashkevich.[109]

IV. RECENT FISSION STUDIES

A. Spontaneous Fission Half-life Systematics

By 1962, a considerable number of SF half-lives had been measured. As shown in Figure 1 these indicated a lengthening of SF half-life for isotopes with 152 neutrons beginning with Z=98 and becoming even more pronounced for fermium and nobelium. Since the half-lives decreased rapidly on either side of N=152, this neutron number was postulated to be a deformed subshell. Because of the perturbation introduced by this subshell the theoretical predictions for nuclei with N greater than 156 were contradictory, some allowing for a recovery of stability even though the experimental trend was pessimistic. Searches for fermium isotopes heavier than mass 257 in fermium chemically separated from the debris of thermonuclear explosions and in reactor-produced fermium were unsuccessful[119,120] even though half-lives of many hours to days were expected for ^{258}Fm and of around a month for ^{259}Fm. The identification in 1971 of ^{258}Fm with a half-life of only 380 μs[57] indicated that a "disaster" in SF half-lives had occurred after N=157. Calculations of Randrup *et al.*[6] indicated that the disappearance of the second fission barrier at ^{258}Fm might account for its very short half-life and thus ^{259}Fm should have about the same half-life except for the special hindrance associated with the odd nucleon. The identification[58] in 1976 of 1.5-second ^{259}Fm was consistent with this explanation and a hindrance factor of 4×10^3 for the 159th neutron. This hindrance to SF from an odd neutron or proton has now been well established[59,60] and is about 10^5 for both odd protons and neutrons although considerable variations have been noted. A number of ideas concerning the origin of this hindrance have been discussed[19] and it seems likely that it is due to the increased fission barrier resulting from conservation of total spin and parity as the nucleus deforms and the odd nucleon cannot transfer to the lowest energy level.

A review of the data for SF by Hoffman and Somerville[59] lists some 118 known SF half-lives. A comparison between experimental and calculated[61] SF half-lives for

the even-even isotopes taken from that review is shown in Figure 5. It can be seen that the stabilization due to the N=152 subshell appears to have disappeared for elements 104 and above, an effect which has made the prediction of SF half-lives for the heaviest elements particularly difficult.

The SF half-life "disaster" at ^{258}Fm coincides with the onset of predominantly symmetric mass division with unusually high $\overline{\text{TKE}}$. This dramatic decrease in half-life has been particularly difficult to calculate theoretically, but recent calculations of Möller *et al.*[62] show the appearance of a new, second valley to fission which leads to configurations close to two touching spheres and is associated with much lower inertia; this treatment gives much better agreement with the shorter half-lives observed for the heavier isotopes. Their calculations still show a considerable shell effect at 152 neutrons for rutherfordium and element 106. Other calculations including reflection-asymmetric shapes[63] also show two fission valleys in the potential-energy surface of heavy Fm isotopes, but further indicate that the new valley lowers the stabilizing effect on SF half-lives of the previously postulated deformed subshell around 162 to 164 neutrons.

Clearly, more experimental information on SF half-lives for still heavier nuclides, and particularly for those with odd numbers of neutrons and/or protons, is essential in order to evaluate the various theoretical models and devise new ones.

B. Spontaneous Fission Properties of Transfermium Isotopes

The mass and kinetic-energy distributions for 8 transfermium isotopes have now been measured[121–122] In another review paper in this volume,[123] a more detailed description of the SF properties of the heavy elements is given.

From the mass-yield curves for the transberkelium isotopes (Figure 3) and the average total kinetic energies of the fission fragments (Figure 4), it can be seen that ^{259}Md appears to be a "transition" nucleus with a broad mass-yield distribution and

a "normal" $\overline{\text{TKE}}$. The transition to a narrowly symmetric mass distribution appears to occur at ^{260}Md, which has a very high $\overline{\text{TKE}}$, similar to those of ^{258}Fm and ^{259}Fm. This transition occurs at one neutron heavier than for Fm. Distributions for four isotopes of nobelium have been measured and these show that ^{256}No is a transition nucleus,[94] but ^{258}No is not narrowly symmetric nor does it show an anomalously high $\overline{\text{TKE}}$. The transition to symmetry thus appears to occur at two or three neutrons heavier in nobelium than in fermium or mendelevium, but the resulting $\overline{\text{TKE}}$ is not anomalously high.

The mass distribution for ^{260}Rf, although symmetric, is extremely broad and its $\overline{\text{TKE}}$ appears somewhat low. The mass distribution for ^{262}Ha has been reported to be asymmetric or very broadly symmetric with a full width at half maximum of 47 mass units or more.[124] At the time of the measurement it seemed unlikely that the mass distribution could be that broad and due to the presence of large amounts of ^{256}Fm (which fissions asymmetrically) it was not possible to make an unequivocal determination. However, its width is not unlike those of ^{257}Fm and ^{256}No.

Hulet *et al.*[121] have decomposed the $\overline{\text{TKE}}$ distributions for ^{258}Fm, ^{259}Md, ^{260}Md, and ^{258}No into two Gaussian distributions of varying intensities, one centered around 200 MeV, and the other around 233 MeV. They call this bimodal fission and suggest that two different fission modes are responsible, a high energy mode arising from compact, spherical scission shapes and a low-energy mode from highly deformed, elongated scission shapes. They attribute the high-energy mode to the fragment shells which are emerging between the saddle and scission point. This becomes less important as the Z and N of the fissioning nuclide moves away from the region where symmetric fission can result in fragments close to the doubly magic (Z=50, N=82), spherical configuration. The low-energy mode may be associated with the disappearance of the second fission barrier predicted to occur in this same general region. This could result in broadly symmetric mass distributions and "normal" $\overline{\text{TKE}}$'s reminiscent of those expected in liquid-drop type fission. These two modes have been

associated with the two valleys to fission discussed in the previous section. However, the "transition" nucleus ^{257}Fm also shows a very large range in TKE's for symmetric mass division, indicating shapes ranging from near-spherical with TKE's approaching the Q value for fission to deformed shapes with low TKE's. This suggests not just two modes but a continuum of shapes ranging from spherical through various combinations of spherical and elongated shapes to predominantly deformed, elongated shapes at scission. Indeed, Paskevich[125] has shown that for ^{264}Fm there are three valleys on the potential-energy surface for symmetric fission in the region of the scission point. One corresponds to a compact configuration of two nearly spherical fragments, another to elongated, more separated fragments, and another to a combination of spherical and elongated fragments. These modes seem to exist in ^{257}Fm, but perhaps there is more of a continuum of shapes rather than just a few modes of fission. A challenging problem which still awaits the theorists is the solution of the dynamic problem involved in determining the probabilities for taking these various paths to fission.

Neutron emission from the fragments has been discussed in other reviews.[59,60] Neutron emission from near spherical, closed-shell fragments with kinetic energies approaching the Q values for fission would be expected to be especially low. This has been verified for ^{257}Fm where fission events with TKE greater than about 240 MeV emit an average of only 1.1 neutrons per fission compared to the overall average value of 3.8; the mass distribution for these high energy events is narrowly symmetric, again indicating near spherical fragments. The heaviest nuclide for which the average number of neutrons per fission has been measured[126] is ^{260}Md. It emits an average of only 2.6 neutrons per fission, a dramatic decrease from the average value for ^{257}Fm. Neutron emission for the higher energy component of the TKE distribution (at 235 MeV) was only 1.8, consistent with the lower neutron emission expected to be associated with near closed-shell fragments with compact shapes. Again, we see the strong influence of shell effects and the extreme sensitivity of the fission process to changes of

only a nucleon or two which makes the development of reliable predictive theoretical models so extremely difficult.

C. Delayed Fission

Delayed fission (DF) is a coupled nuclear decay process in which a decaying nucleus populates excited states in its daughter nucleus, which then fission. Electron capture (EC) or β decay from a parent which has a Q-value smaller than the height of the fission barrier in the daughter should predominantly populate states in the first well, since feeding into the second well would involve a collective rearrangement occurring simultaneously with the EC (or β) decay. Once the high-lying states in the daughter nucleus are populated, tunnelling through the fission barriers must compete with γ transitions for fission to be observed.

Delayed fission was first postulated to influence the production yields of heavy elements in multiple neutron capture processes[64] followed by β decay, such as the astrophysical **r**-process and the production of heavy elements in nuclear devices. Following neutron capture, the neutron-rich nuclei produced in these events begin to β decay towards stability, producing higher atomic number elements. If this chain of β decays were to continue, the very heavy actinides (and possibly superheavy elements) would be produced in amounts that are not observed in nature nor in weapons-tests products.[119,127] It was believed that delayed fission terminates[128–131] the β-decay chain, diminishing the production of the higher Z elements. However, a recent reexamination of weapons-tests data shows that the predicted delayed-fission effects are seriously overestimated.[127,132]

Fission tracks from EC-delayed fission (εDF) were first observed[133,134] in the light americium and neptunium regions as early as 1966. These fissions, however, were originally attributed to a new variant of shape isomerism leading to very long half-lives (on the order of minutes). In 1969, Berlovich and Novikov[135] noted that the

nuclei in question met the theoretical conditions required for delayed fission, although the observed fissions were not specifically attributed[136] by inference to delayed-fission processes until 1972. The observation of fission in ^{232}Am was confirmed by Habs *et al.*[137] in 1978, and the delayed fission probability, P_{DF}, for this isotope was reported to be on the order of one percent (assuming the fissions arose from delayed fission).

Of course, once experimental measurements of delayed-fission branches became available, there were attempts to predict the branches theoretically. In principle, it is possible to extract information about the structure of the daughter's fission barrier from the delayed-fission probability.[137–139] In practice, however, such a calculation is highly dependent on the model used for the feeding of the daughter's levels by the parent's EC or β decay (the β-strength function) and on the amount of damping assumed in the shape isomer. Initial models used empirical approximations for the β-strength function and assumed the shape isomer to be 100% damped (*i. e.*, fission was not allowed to occur prior to γ decay to the lowest state in the second well).[137,139] Subsequent predictions have been based on shell-model calculations of the β-strength function,[131,140] but the problem of damping is not adequately resolved. For calculations very far from stability (which are of astrophysical interest), damping is generally ignored.[131] For nuclei closer to stability (which are experimentally accessible), damping is usually assumed to dominate the second well.[140]

In experimental results, εDF has been reported[141] in nuclei as light as ^{180}Hg, although the majority of observations of delayed fission occur in the actinides. Gangrskiĭ *et al.*[139] report delayed-fission probabilities for several transcurium nuclei using the measured fission ratio to α decay of the EC daughter to estimate σ_ε. Hall *et al.*[142] studied βDF in ^{256}Es^m using radiochemistry and coincidence counting, and identified the excited state in the daughter ^{256}Fm from which fission occurred. Hall *et al.*[143] have performed the first measurements of the time correlation between the electron capture and the subsequent fission in the εDF of ^{234}Am, proving the delayed-fission hypothesis. Hall and coworkers have also used delayed fission to study the near-ground-state

fission properties of ^{232,234}Pu.[144–146] A surprising result in the EC-fission correlation measurement on ^{232,234}Am was the observation of what appear to be discrete γ lines in coincidence with the delayed fission. Hall *et al.* postulated that these lines may arise from γ transitions in the second well,[143–146] although there were insufficient data to attempt to construct a level scheme.

Currently, delayed fission branches have been reported for 13 isotopes. The largest number of reports are based on εDF because of the difficulty in producing very neutron-rich species in a form amenable to study. Of the three nuclides with reported βDF branches (^{256}Es^{m}, ^{236,238}Pa), the reported[147] branch in ^{238}Pa has been recently disproved.[148] This also casts doubt[148] on the report of βDF in ^{236}Pa, since the reported delayed fission in both ^{238}Pa and ^{236}Pa were measured the same way.[147] Based on the work of Baas-May *et al.*,[148] ^{238}Pa has been excluded from the list of delayed-fissioning nuclides in Table I, and it should be noted that the entry for ^{236}Pa is questionable.

V. FUTURE

The future relationship between low-energy fission (of which spontaneous fission is a subset) and our understanding of the atomic nucleus is bound to be varied and complex. We hesitate to make any but the most general predictions, even for the very near future. As in the case of shape isomers, our understanding of the nucleus can be rapidly transformed by totally unexpected experimental results and the subsequent theoretical explanations—or vice-versa. If we could predict what these revolutionary discoveries would be, we would be doing the exciting experiments to make the new discoveries rather than writing this review!

However, it is possible to identify broad areas of fission science that warrant considerable effort in the future. These areas are not easily separated into either theory or experiment since, as we have tried to show in this review, theory and experiment interact in a synergistic manner — with the results of their interaction being greater

than the sum of their individual parts. One such area is the continued investigation of mass-yield and kinetic-energy distributions from low-energy fission. Measurements have been performed on a number of fissioning systems, and in some regions theoretical predictions match the experimental results reasonably well. But the challenge of creating a physically realistic model which can predict the dynamic evolution of the fissioning system, taking account of the structure of both the fissioning nucleus and the fission fragments, and can correctly predict these distributions over the entire range of low-energy fission still remains. Meeting this challenge will require both new theoretical approaches and experimental innovation in order to be able to perform measurements on as many fissioning systems as possible.

Another, somewhat related, area is the problem of SF half-lives. Experimental searches for new nuclides and elements are critically dependent on an ability to predict their half-lives; however, this rather rudimentary quantity has proven highly difficult to model because of its extreme sensitivity to even small changes in the fission barriers. Changes in the barriers as small as 0.1 MeV can lead to changes in SF half-lives on the order of 10^5. Thus, meaningful half-life calculations require a remarkably complete understanding of nuclear structure, the effect of odd nucleons, and details of the path to scission.

Many experiments and theories will fall into these broad areas, but many others undoubtedly will not. Such is the nature of scientific research and progress. However, one can be rather sure that the continued interplay of theory and experiment will provide many intriguing new results and will give new insights into our understanding of matter on the subatomic scale.

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[1]E. FERMI, *Nature 133* (1934) 898.

[2]O. HAHN and F. STRASSMANN, *Naturwissenschaften 27* (1939) 11.

[3]O. HAHN and F. STRASSMANN, *Naturwissenschaften 27* (1939) 89.

[4]L. MEITNER and O. R. FRISCH, *Nature 143* (1939) 239.

[5]N. BOHR, *Nature 137* (1936) 344.

[6]J. RANDRUP, C. F. TSANG, P. MÖLLER, S. G. NILSSON, S. E. LARSSON, *Nucl. Phys. A217* (1973) 221.

[7]G. N. FLEROV and K. A. PETRZHAK, *Phys. Rev. 58* (1940) 89. G. N. FLEROV and K. A. PETRZHAK, *J. Phys. (USSR) 3* (1940) 275.

[8]N BOHR and J. R. WHEELER, *Phys. Rev. 56* (1939) 426.

[9]H. R. VON GUNTEN, *Actinides Rev. 1* (1969) 275.

[10]M. G. MAYER, *Phys. Rev. 78* (1950) 16.

[11]O. HAXEL, J. H. D. JENSEN, and H. E. SEUSS, *Z. Phys. 128* (1950) 295.

[12]M. G. MAYER and J. H. D. JENSEN, **Elementary Theory of Nuclear Shell Structure** (Wiley, New York 1955).

[13]K. F. FLYNN, E. P. HOROWITZ, C. A. A. BLOOMQUIST, R. F. BARNERS, R. K. SJOBLOM, R. R. FIELD, and L. E. GLENDENIN, *Phys. Rev. C 5* (1972) 1725.

[14]I. KELSON, *Phys. Rev. Lett. 20* (1968) 867.]

[15]J. A. FARRELL, *Phys. Rev. 165* (1968) 1371.

[16]A. BOHR, *in* **Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955, Volume 2** (United Nations, New York 1956) 151.

[17]G. A. COWAN, B. P. BAYHURST, R. J. PRESTWOOD, J. S. GILMORE, and G. W. KNOBELOCH, *Phys. Rev. 144* (1966) 197.

[18]E. MELKONIAN and G. K. MEHTA, *in* **Proceedings of the IAEA Symposium on the Physics and Chemistry of Fission, Salzburg, 1965, Volume 2**, (IAEA, Vienna 1965) 355.

[19]R. VANDENBOSCH and J. R. HUIZENGA, **Nuclear Fission**, Academic Press (New York, 1973).

[20]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, and V. A. FOMICHEV, *Zh. Eksp. Teor. Fiz. 42* (1962) 1464 [*Sov. Phys. JETP 15*, (1962) 1016].

[21]V. P. PERELYGIN, S. P. ALMAZOVA, B. A. GVOZDEV, and YU. T. CHUBERKOV, *Zh. Eksp. Teor. Fiz. 42* (1962) 1472 [*Sov. Phys. JETP 15*, (1962) 1022].

[22]A. GHIORSO, *in* **Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955, Volume 7** (United Nations, New York 1955), 15.

[23]V. A. KARNAUKHOV, I. KHALIZEV, and G. N. FLEROV, *Zh. Eksp. Teor. Fiz. 37* (1959) 1266 [*Sov. Phys. JETP 10*, (1960) 901].

[24]V. A. KARNAUKHOV, *Zh. Eksp. Teor. Fiz. 42* (1962) 973 [*Sov. Phys. JETP 15*, (1962) 671].

[25]H. C. BRITT, *At. Data Nucl. Data Tables 12*, (1973) 407.

[26]V. M. STRUTINSKY, *Nucl. Phys. A95* (1967) 420.

[27]V. M. STRUTINSKY, *Nucl. Phys. A122* (1968) 1.

[28]S. BJØRNHOLM, J. BORGGREEN, L. WESTGAARD, and V. A. KARNAUKHOV, *Nucl. Phys. A95* (1967) 513.

[29]G. N. FLEROV, YU. P. GANGRSKIĬ, B. N. MARKOV, A. A. PLEVE, S. M. POLIKANOV, and KH. YUNGKLAUSSEN, *Yad. Fiz. 6* (1967) 17 [*Sov. J. Nucl. Phys. 6* (1968) 12].

[30]YU. P. GANGRSKIĬ, K. A. GAVRILOV, B. N. MARKOV, NGUYEN KONG KHAN, and S. M. POLIKANOV, *Yad. Fiz. 10* (1969) 65 [*Sov. J. Nucl. Phys. 10* (1970) 38].

[31]N. L. LARK, G. SLETTEN, J. PEDERSEN, and S. BJØRNHOLM, *Nucl. Phys. A139* (1969) 481.

[32]S. M. POLIKANOV and G. SLETTEN, *Nucl. Phys. A151* (1970) 656.

[33]H. J. SPECHT, J. WEBER, E. KONECNY, and D. HEUNEMANN, *Phys. Lett. B 41* (1972) 43.

[34]D. N. POENARU, M. S. IVAŞCU, and D. MAZILU, *in* **Charged Particle Emission from Nuclei Vol. III**, D. N. POENARU and M. IVAŞCU, eds., (CRC Press, Boca Raton, Fla., 1989) 41.

[35]J. CHRISTIANSEN, G. HEMPEL, H. INGWERSEN, W. KLINGER, G. SCHATZ,

and W. WITTHUHN, *Nucl. Phys. A239* (1975) 253.

[36]J. DREXLER, R. D. HEIL, K. HUBER, U. KNIESSL, G. MANK, R. RATZEK, H. REIS, T. WEBER, W. WILKE, B. FISCHER, and H. HOLLICK, *Nucl. Phys. A437* (1985) 253.

[37]S. BURNETT, H. BRITT, B. ERKKILA, and W. STEIN, *Phys. Lett. B 31* (1970) 523.

[38]R. REPNOV, V. METAG, and P. VON BRENTANO, *Z. Phys. 243* (1971) 419.

[39]V. METAG, D. HABS, and H. J. SPRECHT, *Phys. Rep. 65* (1980) 1.

[40]S. DE BARROS, S. D. DE MAGALHÃES, H. WOLF, J. BARRETO, J. EICHLER, N. LISBONA, I. O. DE SOUZA, and D. M. VIANNA, *Z. Physik A323* (1986) 101.

[41]V. METAG, E. LIUKKONEN, O. GLOMSET, and A. BERGMAN, *in* **Proceedings of the IAEA Symposium on Physics and Chemistry of Fission** (IAEA, Vienna, 1974) 317.

[42]S. BJØRNHOLM and J. E. LYNN, *Rev. Mod. Phys. 52* (1980) 725.

[43]K. L. WOLF and J. P. UNIK, *Phys. Lett. B 38*, (1972) 405.

[44]G. N. FLEROV, A. PLEVE, S. POLIKANOV, S. TRETYAKOVA, N. Martalogu, D. POENARU, M. SEZON, I. VILCOV, and N. VILCOV, *Nucl. Phys. A97* (1967) 444.

[45]S. BJØRNHOLM, I. BORGGREEN, YU. GANGRSKIĬ, and G. SLETTEN, *Yad. Fiz. 8* (1968) 459 [*Sov. J. Nucl. Phys. 8* (1969) 267].

[46]G. SLETTEN, V. METAG, and E. LIUKKONEN, *Phys. Lett. B 60* (1976) 153.

[47]H. C. BRITT, S. BURNETT, B. ERKKILA, J. LYNN, and W. STEIN, *Phys. Rev. C 4* (1971) 1441.

[48]W. D. MYERS and W. J. ŚWIĄTECKI, *Nucl. Phys. 81* (1966) 1.

[49]H. W. MELDNER, *Ark. Fys. 36* (1967) 593.

[50]S. G. NILSSON, C. F. TSANG, A. SOBICZEWSKI, Z. SZYMAŃSKI, S. WYCECH, C. GUSTAFSON, I.-L. LAMM, P. MÖLLER, and B. NILSSON, *Nucl. Phys. A131* (1969) 1.

[51]G. T. SEABORG, *Phys. Scripta 10A* (1974) 5.

[52]O. L. KELLER and G. T. SEABORG, *Ann. Rev. Nucl. Sci. 27* (1977) 139.

[53]V. M. VDOVENKO and E. V. SOBOTOVICH, *Dokl. Akad. Nauk. SSSR 189* (1969) 980 [*Sov. Phys. Doklady 14* (1969) 1179].

[54]U. HAACK, *Naturwissenschaften 60* (1973) 65.

[55]P. R. FIELDS, H. DIAMOND, D. N. METTA, C. M. STEVENS, D. J. ROKOP, and P. E. MORELAND, *Science 167*, (1970) 499.

[56]E. CHEIFETZ, R. C. JARED, E. R. GIUSTI, and S. G. THOMPSON, *Phys. Rev. C 6* (1972) 1348.

[57]E. K. HULET, J. F. WILD, R. W. LOUGHEED., J. E. EVANS, B. J. QUALHEIM, M. NURMIA, and A. GHIORSO, *Phys. Rev. Lett. 26* (1971) 523.

[58]D. C. HOFFMAN, J. WEBER, J. B. WILHELMY, E. K. HULET, J. H. LANDRUM, R. W. LOUGHEED, and J. F. WILD, *in* **Proceedings of the Third International Conference on Nuclei Far from Stability, Cargese, Corsica, France,** (CERN, Geneva 1976) 558.

[59]D. C. HOFFMAN and L. P. Somerville, *in* **Charged Particle Emission from Nuclei Vol. III**, D. N. POENARU and M. Ivascu, eds., (CRC Press, Boca Raton, Fla., 1989) 1.

[60]D. C. HOFFMAN, *in* **Proceedings of the Conference on Fifty Years with**

Nuclear Fission, Vol. I (American Nuclear Society, La Grange Park, Ill. 1989) 83.

[61]J. RANDRUP, S. E. LARSSON, P. MÖLLER, S. G. NILSSON, K. POMORSKI, and A. SOBICZEWSKI, *Phys. Rev. C 13* (1976) 229.

[62]P. MÖLLER, J. R. NIX, and W. J. ŚWIĄTECKI, *Nucl. Phys. A492* (1989) 349.

[63]S. CWIOK, P. ROZMEJ, A. SOBICZEWSKI, and Z. PATYK, *Nucl. Phys. A491* (1989) 281.

[64]E. M. BURBIDGE, G. R. BURBIDGE, W. A. FOWLER, and F. HOYLE, *Rev. Mod. Phys. 29* (1957) 547.

[65]P. H. FOWLER, V. M. CLAPHAM, V. G. COWEN, J. M. KIDD, and R. T. MOSES, *Proc. Roy. Soc. (London) A318* (1970) 1.

[66]P. B. PRICE, P. H. FOWLER, J. M. KIDD, E. J. KOBETICH, R. L. FLEISCHER, and G. E. NICHOLS, *Phys. Rev. D 3* (1971) 815.

[67]O. OTGONSURÉN and V. P. PERELYGIN, *Joint Institute for Nuclear Research, Dubna, Report No. JINR P7-7406* (1973).

[68]W. GRIMM, G. HERRMANN, and H.-D. SCHÜSSLER, *Phys. Rev. Lett. 26* (1971) 1040.

[69]O. OTGONSURÉN, V. P. PERELYGIN, and G. N. FLEROV, *Dokl. Akad. Nauk. SSSR 189* (1969) 1200 [*Sov. Phys. Doklady 14* (1969) 1194].

[70]H. R. BOWMAN, R. C. GATTI, R. C. JARED, L. G. MORETTO, W. J. ŚWIĄTECKI, and S. G. THOMPSON, *Lawrence Berkeley Laboratory Report No. UCRL-17969* (1967).

[71]P. COLOMBANI, B. GATTY, J. C. JACMART, M. LEFORT, J. PETER, M. RIOU, C. STEPHEN, and X. TARRAGO, *Phys. Lett. B 42* (1972) 208.

[72]A. PLEVE, A. G. DEMIN, V. KUSH, M. B. MILLER, and N. A. DANILOV, *Yad. Fiz. 19* (1974) 252 [*Sov. J. Nucl. Phys. 19* (1974) 123].

[73]M. BOLSTERLI, E. O. FISET, J. R. NIX, and J. L. NORTON, *Phys. Rev. C5* (1972) 1050. Also, *Phys. Rev. Lett. 27* (1971) 61.

[74]E. O. FISET and J. R. NIX, *Nucl. Phys. A193* (1972) 649.

[75]M. BRACK, J. DAMGAARD, A. S. JENSES, H. C. PAULI, V. M. STRUTINSKY, and C. Y. WONG, *Rev. Mod. Phys. 44* (1972) 185.

[76]E. K. HULET, R. W. LOUGHEED, J. F. WILD, J. H. LANDRUM, P. C. STEVENSON, A. GHIORSO, J. M. NITSCHKE, R. J. OTTO, D. J. MORRISSEY, P. A. BAISDEN, B. F. GAVIN, D. LEE, R. J. SILVA, M. M. FOWLER, and G. T. SEABORG, *Phys. Rev. Lett. 39* (1977) 385.

[77]J. D. ILLIGE, E. K. HULET, J. M. NITSCHKE, R. T. DOUGAN, R. W. LOUGHEED, A. GHIORSO, and J. H. LANDRUM, *Phys. Lett. B 78* (1978) 209.

[78]YU. TS. OGANESSIAN, H. BRUCHERTSEIFER, G. V. BUKLANOV, V. I. CHEPIGIN, CHOI VAL SEK, B. EICHLER, K. A. GAVRILOV, H. GÄGGELER, YU. S. KOROTKIN, O. A. ORLOVA, T. REETZ, W. SEIDEL, G. M. TER-AKOPIAN, S. P. TRETYAKOVA, and I. ZVARA, *Nucl. Phys. A294* (1978) 213.

[79]R. J. OTTO, D. J. MORRISSEY, D. LEE, A. GHIORSO, J. M. NITSCHKE, and G. T. SEABORG, *J. Inorg. Nucl. Chem. 40* (1978) 589.

[80]P. ARMBRUSTER, Y. K. AGARWAL, W. BRÜCHLE, M. BRÜGGER, J. P. DUFOUR, H. GÄGGELER, F. P. HESSBERGER, S. HOFMANN, P. LEMMERTZ, G. MÜNZENBERG, K. POPPENSIEKER, W. REISDORF, M. SCHÄDEL, K.-H. SCHMIDT, J. H. R. SCHNEIDER, W. F. W. SCHNEIDER, K. SÜMMERER,

D. VERMEULEN, G. WIRTH, A. GHIORSO, K. E. GREGORICH, D. LEE, M. LEINO, K. J. MOODY, G. T. SEABORG, R. B. WELCH, P. WILMARTH, S. YASHITA, C. FRINK, N. GREULICH, G. HERRMANN, U. HICKMANN, N. HILDEBRAND, J. V. KRATZ, N. TRAUTMANN, M. M. FOWLER, D. C. HOFFMAN, W. R. DANIELS, H. R. VON GUNTEN, and H. DORNHÖFER, *Phys. Rev. Lett. 54*, (1985) 406.

[81]H. GÄGGELER, N. TRAUTMANN, W. BRÜCHLE, G. HERRMANN, J. V. KRATZ, P. PEUSER, M. SCHÄDEL, G. TITTEL, G. WIRTH, H. AHRENS, H. FOLGER, G. FRANZ, K. SÜMMERER, and M. ZENDEL, *Phys. Rev. Lett. 45* (1980) 1824.

[82]H. GÄGGELER, D. JOST, A. TÜRLER, P. ARMBRUSTER, W. BRÜCHLE, H. FOLGER, F. P. HESSBERGER, S. HOFMANN, G. MÜNZENBERG, V. NINOV, M. SCHÄDEL, K. SÜMMERER, J. V. KRATZ, U. SCHERER, and M. LEINO, *Proceeding of the International Conference on Fifty Years of Research in Nuclear Fission, Berlin, April 3–7, 1989* (in press).

[83]M. LEINO, S. YASHITA, and A. GHIORSO, *Phys. Rev. C 24*, (1981) 2370.

[84]G. MÜNZENBERG, W. FAUST, S. HOFMANN, P. ARMBRUSTER, K. GÜTTNER, and H. EWALD, *Nucl. Inst. Meth. 161* (1979) 65.

[85]G. T. SEABORG, W. LOVELAND, and D. J. MORRISSEY, *Science 203* (1979) 711.

[86]G. T. SEABORG and W. LOVELAND, *Contemp. Phys. 28* (1987) 33.

[87]G. HERRMANN, *Proceedings of the International Symposium on Artificial Radioactivity, Poona, January 8–12, 1985.* Also, *Gessellschaft für Schwerionenforschung, Darmstadt Report Number GSI-85-10.*

[88]P. ARMBRUSTER, *Ann. Rev. Nucl. Part. Science, 35* (1986) 135.

[89]P. ARMBRUSTER and G. MÜNZENBERG, *Scientific American*, 66 (May 1989).

[90]A. GHIORSO, D. C. HOFFMAN, E. K. HULET, O. L. KELLER, and G. T. SEABORG, *Lawrence Berkeley Laboratory Publication Number PUB-5118* (1984).

[91]J. P. BALAGNA, G. P. FORD, D. C. HOFFMAN, and J. D. KNIGHT, *Abstracts, 159th National Meeting of the American Chemical Society, Houston* (1970).

[92]J. P. BALAGNA, G. P. FORD, D. C. HOFFMAN, and J. D. KNIGHT, *Phys. Rev. Lett. 26* (1971) 145.

[93]W. JOHN, E. K. HULET, R. W. LOUGHEED, and J. J. WESOLOWSKI, *Phys. Rev. Lett. 27* (1971) 45.

[94]D. C. HOFFMAN, D. M. LEE, K. E. GREGORICH, M. J. NURMIA, R. B. CHADWICK, K. B. CHEN, K. R. CZERWINSKI, C. M. GANNETT, H. L. HALL, R. A. HENDERSON, B. KADKHODAYAN, S. A. KREEK, and J. D. LEYBA, *Phys. Rev. C* (1990) in press. Also, *Lawrence Berkeley Laboratory Report No. LBL-27667* (1989).

[95]V. VIOLA, *Nucl. Data, Sect. B 1* (1966) 391.

[96]J. P. UNIK, J. E. GINDLER, L. E. GLENDENIN, K. F. FLYNN, A. GORSKI, and R. K. SJOBLOM, *in* **Proceedings of the 3rd IAEA Symposium on the Physics and Chemistry of Fission, 1973, Vol. 2** (International Atomic Energy Agency, Vienna, 1974), 19.

[97]D. C. HOFFMAN, *in* **Proceedings of the Third International Conference on Nuclei Far From Stability, Cargese, Corsica, France, 1976** (CERN, Geneva 1976) 565.

[98]D. C. HOFFMAN, J. B. WILHELMY, J. WEBER, W. R. DANIELS, E. K. HULET, R. W. LOUGHEED, J. H. LANDRUM, J. F. WILD, and

R. J. DUPZYK, *Phys. Rev. C 21* (1980) 972.

[99]V. M. STRUTINSKY, *in* **Proceedings of the International Symposium on Why and How to Investigate Nuclides Far Off the Stability Line, Lysekil, 1966**, *Ark. Fys. 36* (1966) 629.

[100]P. MÖLLER and S. G. NILSSON, *Phys. Lett. 31B* (1970) 283.

[101]J. R. NIX, *Los Alamos Scientific Laboratory Report No. LA-DC-72-335* (1972).

[102]J. R. NIX, *Los Alamos Scientific Laboratory Report No. LA-DC-72-769* (1972).

[103]C. F. TSANG and J. B. WILHELMY, *Nucl. Phys. A184* (1972) 417.

[104]M. G. MUSTAFA, U. MOSEL, and H. W. SCHMITT, *Phys. Rev. Lett. 28* (1972) 1536.

[105]M. G. MUSTAFA, U. MOSEL, and H. W. SCHMITT, *Phys. Rev. C 7* (1972) 1519.

[106]B. D. WILKINS, E. P. STEINBERG, and R. R. CHASMAN, *Phys. Rev. C 14* (1973) 1832.

[107]A. W. FAIRHALL, *Phys. Rev. 102* (1956) 1335.

[108]R. C. JENSEN and A. W. FAIRHALL, *Phys. Rev. 109* (1958) 942.

[109]V. V. PASHKEVICH, *Nucl. Phys. A169* (1971) 275.

[110]V. V. PASHKEVICH, *in* **International Seminar School on Heavy Ion Physics** (JINR, Dubna, 1983) 405.

[111]A. TURKEVICH and J. B. NIDAY, *Phys. Rev. 84*, (1951) 52.

[112]T. T. SUGIHARA, J. ROESMER, and J. W. MEADOWS, JR., *Phys. Rev. 121* (1961) 1179.

[113]F. PLASIL, R. L. FERGUSON, F. PLEASONTON, and H. W. SCHMITT, *Phys. Rev. C 7* (1973) 1186.

[114]E. N. GRUZINTSEV, M. G. ITKIS, V. N. OKLOVICH, A. YA. RUSANOV, G. N. SMIRENKIN, and V. N. TOLSTIKOV, *Pis'ma Zh. Eksp. Teor. Fiz. 35* (1982) 449 [*JETP Lett. 35* (1983) 557].

[115]E. N. GRUZINTSEV, M. G. ITKIS, V. N. OKLOVICH, A. YA. RUSANOV, G. N. SMIRENKIN, and V. N. TOLSTIKOV, *Pis'ma Zh. Eksp. Teor. Fiz. 36* (1982) 304 [*JETP Lett. 36* (1983) 372].

[116]M. G. ITKIS, V. N. OKLOVICH, A. YA. RUSANOV, and G. N. SMIRENKIN, *Yad. Fiz. 39* (1984) 1349 [*Sov. J. Nucl. Phys. 39* (1985) 852].

[117]M. G. ITKIS, V. N. OKLOVICH, A. YA. RUSANOV, and G. N. SMIRENKIN, *Yad. Fiz. 41* (1985) 849 [*Sov. J. Nucl. Phys. 41*, (1985) 544].

[118]M. G. ITKIS, V. N. OKLOVICH, and A. YA. RUSANOV, *Fiz. Elem. Chastits At. Yadra 19* (1988) 701 [*Sov. J. Part. Nucl. 19* (1988) 301].

[119]COMBINED RADIOCHEMISTRY GROUP, *Phys. Rev. 148*, (1966) 1192.

[120]D. C. HOFFMAN, *in* **Proceedings of the International Symposium on Why and How to Investigate Nuclides Far Off the Stability Line, Lysekil, 1966**, *Ark. Fys. 36* (1966) 533.

[121]E. K. HULET, J. F. WILD, R. J. DOUGAN, R. W. LOUGHEED, J. H. LANDRUM, A. D. DOUGAN, P. A. BAISDEN, C. M. HENDERSON, R. J. DUPZYK, R. L. HAHN, M. SCHÄDEL, K. SÜMMERER, and G. R. BETHUNE, *Phys. Rev. C 40* (1989) 770.

[122]R. W. LOUGHEED, E. K. HULET, J. F. WILD, K. J. MOODY, R. J. DOUGAN, C. M. GANNETT, R. A. HENDERSON, D. C. HOFFMAN, and D. M. LEE, "Discovery of 5-ms ^{262}No and the Study of its Spontaneous-Fission Decay Properties," *Nuclear Chemistry Division FY 1988 Annual Report, Lawrence Livermore National Laboratory Report No. UCAR 10062-88* (1988) 132.

[123]E. K. HULET, "Spontaneous Fission in the Heavy Elements," this volume. Also, *Lawrence Livermore National Laboratory Report No. UCRL-101880* (1989).

[124]C. E. BEMIS, JR., R. L. FERGUSON, F. PLASIL, R. J. SILVA, G. D. O'KELLEY, M. L. KIEFER, R. L. HAHN, D. C. HENSLEY, E. K. HULET, R. W. LOUGHEED, *Phys. Rev. Lett. 39* (1977) 1246.

[125]V. V. PASKEVICH, *Nucl. Phys. A477* (1988) 1.

[126]J. F. WILD, J. VAN AARLE, W. WESTMEIER, R. W. LOUGHEED, E. K. HULET, K. J. MOODY, R. J. DOUGAN, R. BRANDT, and P. PATZELT, *Phys. Rev. C* (1990) in press. Also, "Neutron Emission as a Function of Fragment Energy in the Spontaneous Fission of ^{260}Md," *Nuclear Chemistry Division FY 1988 Annual Report, Lawrence Livermore National Laboratory Report No. UCAR 10062-88* (1988) 132.

[127]R. W. HOFF, *in* **Weak and Electromagnetic Interactions in Nuclei**, H. V. KLAPDOR, ed., Springer-Verlag (Heidelburg, 1986), 207.

[128]C.-O. WENE and S. A. E. JOHANSSON, *Phys. Scripta 10A* (1974) 156.

[129]C.-O. WENE, *Astron. & Astrophys. 44* (1975) 233.

[130]H. V. KLAPDOR, T. ODA, J. METZINGER, W. HILLEBRANDT, and F. K. THIELMAN, *Z. Physik A 299* (1981) 213.

[131]B. S. MEYER, W. M. HOWARD, G. J. MATTHEWS, K. TAKAHASHI, P. MÖLLER, and G. LEANDER, *Phys. Rev. C 39* (1989) 1876.

[132]R. W. HOFF, *Inst. Phys. Conf. Ser. No. 88/J. Phys. G:Nucl. Phys. 14 Suppl.* (1986) S343.

[133]V. I. KUZNETSOV, N. K. SKOBELEV, and G. N. FLEROV, *Yad. Fiz. 4* (1966) 279 [*Sov. J. Nucl. Phys. 4* (1967) 202].

[134]V. I. KUZNETSOV, N. K. SKOBELEV, and G. N. FLEROV, *Yad. Fiz. 5* (1967) 271 [*Sov. J. Nucl. Phys. 5* (1967) 191].

[135]É. E. BERLOVICH and YU. P. NOVIKOV, *Dok. Akad. Nauk SSSR 185* (1969) 1025 [*Sov. Physics—Doklady 14* (1969) 349].

[136]N. K. SKOBELEV, *Yad. Fiz. 15* (1972) 444 [*Sov. J. Nucl. Phys. 15* (1972) 249].

[137]D. HABS, H. KLEWE-NEBENIUS, V. METAG, B. NEUMANN, and H. J. SPECHT, *Z. Physik A 285* (1978) 53.

[138]Yu. A. LAZAREV, YU. TS. OGANESSIAN, and V. I. KUZNETSOV, *Joint Institutes for Nuclear Research, Dubna, Report No. JINR-E7-80-719* (1980).

[139]YU. P. GANGRSKIĬ, M. B. MILLER, L. V. MIKHAĬLOV, and I. F. KHARISOV, *Yad. Fiz. 31* (1980) 306 [*Sov. J. Nucl. Phys. 31* (1980) 162].

[140]H. V. KLAPDOR, C.-O. WENE, I. N. ISOSIMOV, and YU. W. NAUMOW, *Z. Physik A 292* (1979) 249.

[141]Yu. A. LAZAREV, YU. TS. OGANESSIAN, I. V. SHIROKOVSKY, S. P. TRETYAKOVA, V. K. UTYONKOV, and G. V. BUKLANOV, *Europhys. Lett. 4* (1987) 893.

[142]H. L. HALL, K. E. GREGORICH, R. A. HENDERSON, D. M. LEE, D. C. HOFFMAN, M. E. BUNKER, M. M. FOWLER, P. LYSAGHT, J. W. STARNER, and J. B. WILHELMY, *Phys. Rev. C 39* (1989) 1866.

[143]H. L. HALL, K. E. GREGORICH, R. A. HENDERSON, C. M. GANNETT, R. B. CHADWICK, J. D. LEYBA, K. R. CZERWINSKI, B. KADKHODAYAN, S. A. KREEK, D. M. LEE, M. J. NURMIA, and D. C. HOFFMAN, *Phys. Rev. Lett. 63* (1989) 2548.

[144]H. L. HALL, K. E. GREGORICH, R. A. HENDERSON, C. M. GANNETT,

R. B. CHADWICK, J. D. LEYBA, K. R. CZERWINSKI, B. KADKHODAYAN, S. A. KREEK, D. M. LEE, M. J. NURMIA, D. C. HOFFMAN, C. E. A. PALMER, and P. A. BAISDEN, *Phys. Rev. C* (1989) in press. Also, *Lawrence Berkeley Laboratory Report No. 27510* (1989).

[145]H. L. HALL and D. C. HOFFMAN, *in* **Proceedings of the American Chemical Society Symposium on Exotic Nuclear Spectroscopy, Miami Beach 1989**, W. C. MCHARRIS, ed., (*in press*). Also, *Lawrence Berkeley Laboratory Report No. LBL-27831* (1989).

[146]H. L. HALL, **Delayed-Fission Properties of Neutron-Deficient Americium Nuclei** (Ph. D. Thesis, University of California, 1989). Also, *Lawrence Berkeley Laboratory Report No. LBL-27878* (1989).

[147]Yu. P. GANGRSKIĬ, G. M. MARINESCU, M. B. MILLER, V. N. SAMOYUSK, and I. F. KHARISOV, *Yad. Fiz. 27* (1978) 894 [*Sov. J. Nucl. Phys. 27* (1978) 475].

[148]A. BAAS-MAY, J. V. KRATZ, and N. TRAUTMANN, *Z. Physik A 322* (1985) 457.

[149]R. HINGMAN, W. KUEHN, V. METAG, R. NOVOTNY, A. RUCKELSHAUSEN, H. STROEHER, F. HESSBERGER, S. HOFMANN, G. MUENZENBERG, and W. REISDORF, *Gessellschaft für Schwerionenforschung, Darmstadt, Report No. GSI 85-1*, 88 (1985).

[150]L. P. SOMERVILLE, A. GHIORSO, M. J. NURMIA, and G. T. SEABORG, *Lawrence Berkeley Laboratory Nuclear Science Division Annual Report, 1976-1977, Report No. LBL-6575* (1977) 39.

TABLE I. Summary of reported observations of delayed fission.

Nuclide[a]	$t_{1/2}$	P_{DF}	Reference
^{250}Md (εDF)	52 sec.	2×10^{-4}	Gangrskiĭ[139]
^{256m}Es (βDF)	7.6 hour	2×10^{-5}	Hall[142]
^{248}Es (εDF)	28 min.	3×10^{-7}	Gangrskiĭ[139]
^{246}Es (εDF)	8 min.	3×10^{-5}	Gangrskiĭ[139]
^{244}Es (εDF)	37 sec.	10^{-4}	Gangrskiĭ[139]
^{242}Es (εDF)	5 - 25 sec.	$(1.4 \pm 0.8) \times 10^{-2}$	Hingmann[149]
^{240}Bk (εDF)	4 min.	10^{-5}	Gangrskiĭ[139]
^{234}Am (εDF)	2.6 ± 0.2 min.	Not Reported (NR)	Skobelev[136]
^{234}Am (εDF)	2.6 ± 0.2 min.	NR	Somerville[150]
^{234}Am (εDF)	2.32 ± 0.08 min.	$(6.6 \pm 1.8) \times 10^{-5}$	Hall[144,146]
^{232}Am (εDF)	1.4 ± 0.25 min.	NR	Skobelev[136]
^{232}Am (εDF)	0.92 ± 0.12 min.	$1.3^{+4}_{-0.8} \times 10^{-2}$	Habs[137]
^{232}Am (εDF)	1.31 ± 0.04 min.	$(6.9 \pm 1.0) \times 10^{-4}$	Hall[145,146]
^{228}Np (εDF)	60 ± 5 sec.	NR	Skobelev[136]
^{236}Pa? (βDF)	9.1 min.	3×10^{-10}	Gangrskiĭ[147]
^{180}Tl? (εDF)	$0.70^{+0.12}_{-0.09}$ sec.	$\sim 10^{-6}$	Lazarev[141]

[a]The parent nuclide undergoing decay to a daughter which then fissions is given.

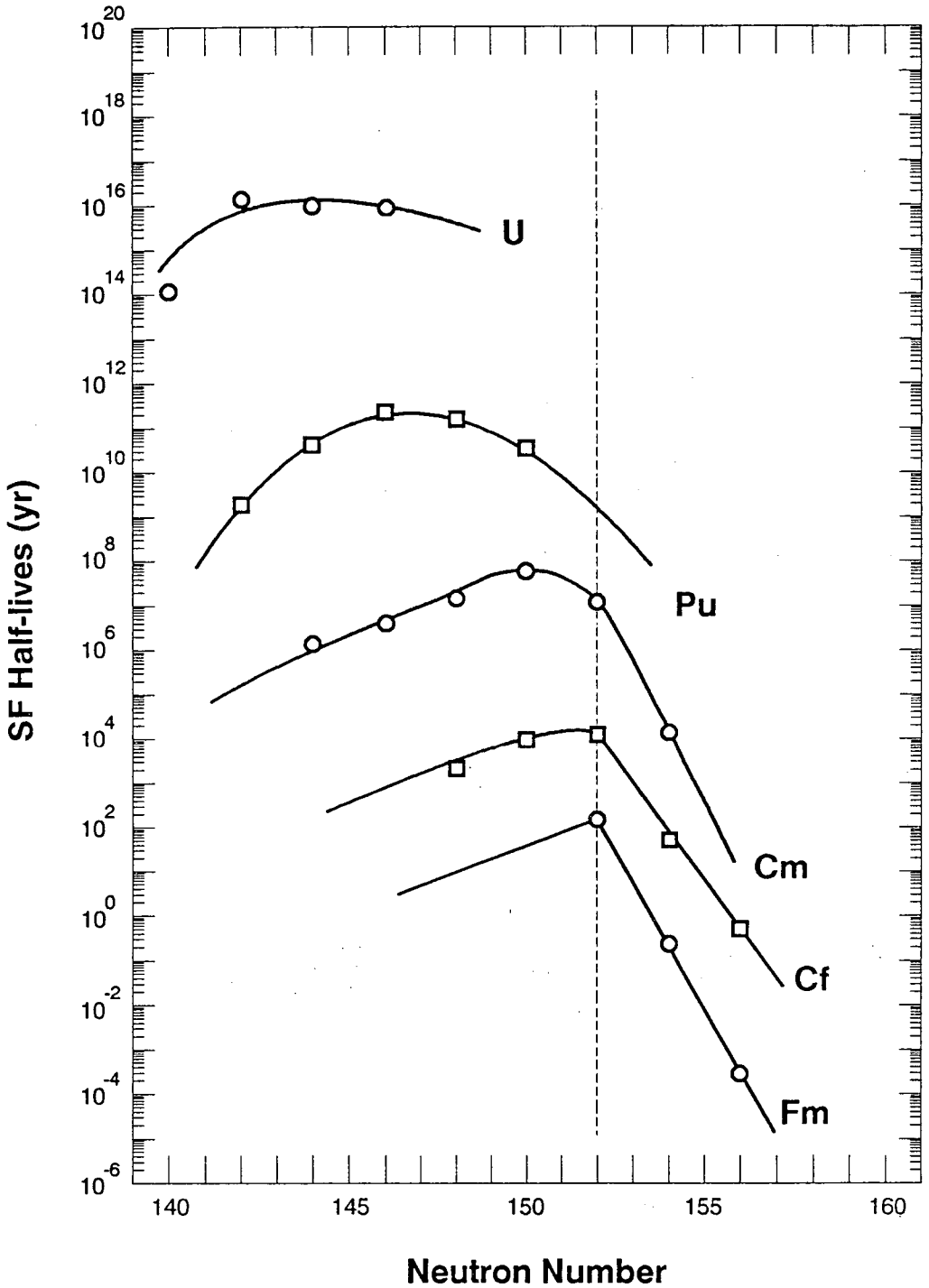
FIG. 1. Fission half-lives known in 1962 as a function of neutron number. After Ghiorso.[22]

FIG. 2. Nuclear potential energy as a function of deformation.

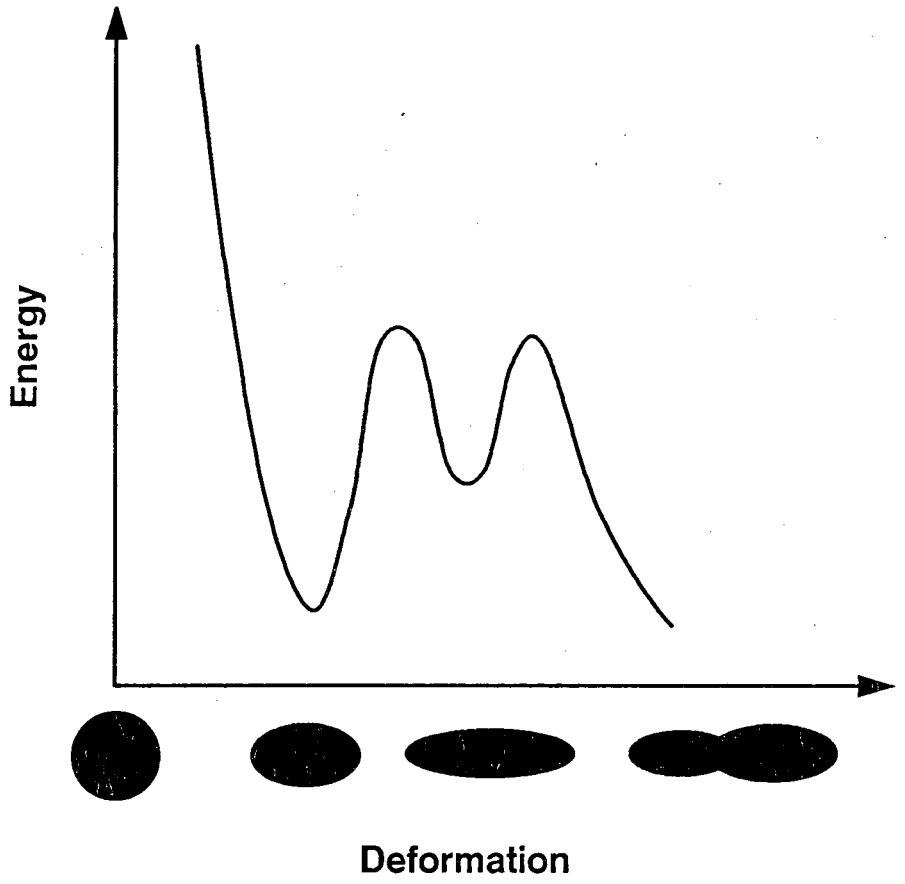
FIG. 3. Schematic representation of mass-yield distributions for transberkelium isotopes as of 1989,[94] normalized to 200% fission fragment yield.

FIG. 4. Total kinetic energy released in fission (TKE) as a function of $Z^2/A^{1/3}$. The solid line is a linear fit of Viola,[95] and the dashed line is from Unik *et al.*[96] Solid squares (■) indicate data from spontaneous fission, while the solid circles (•) indicate data from delayed fission.

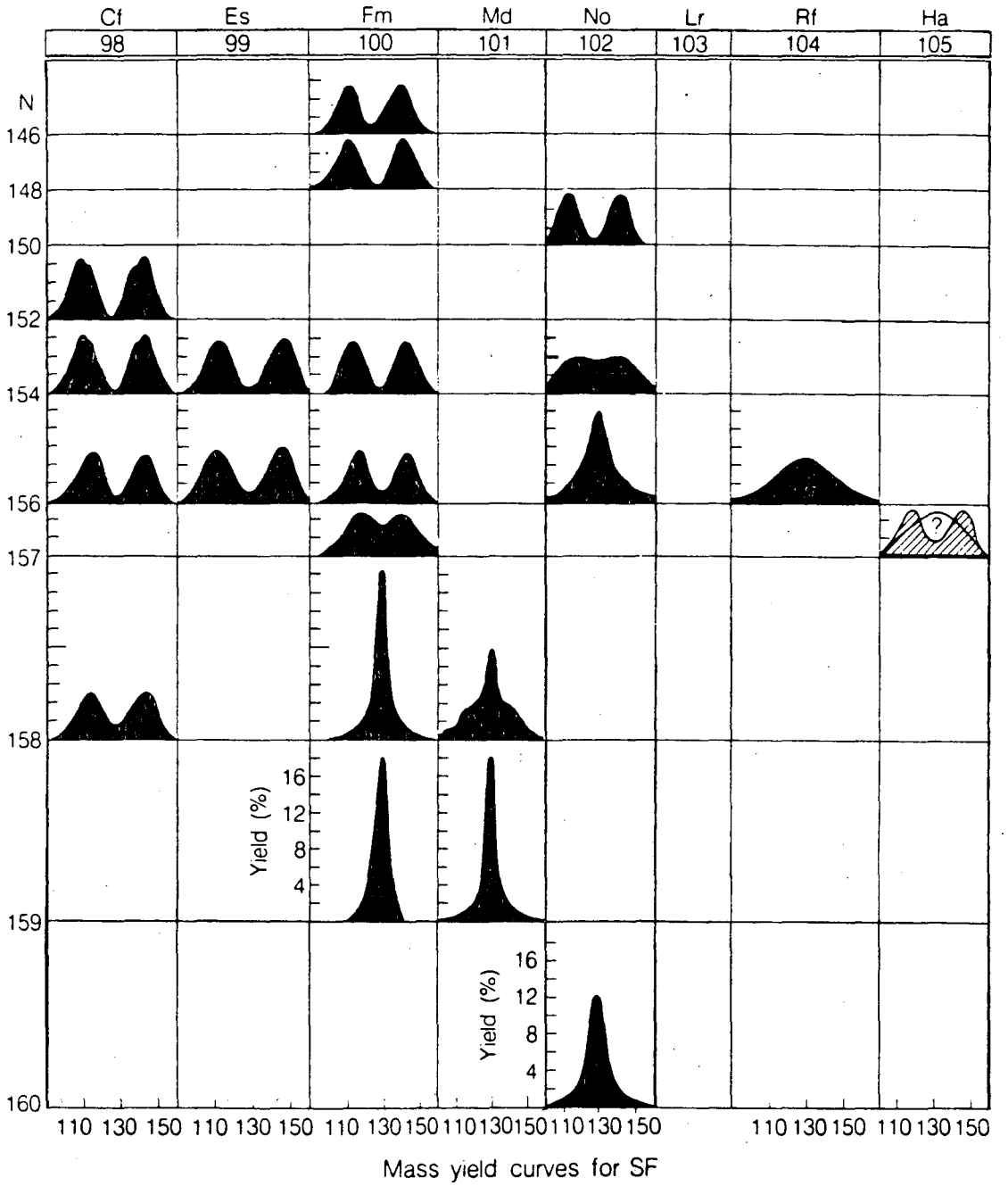
FIG. 5. Experimental and calculated[61] spontaneous fission half-lives. (Figure is taken from the review by Hoffman and Somerville,[59] and reprinted with permission.)



Hall & Hoffman – Fig. 1



Hall & Hoffman – Fig. 2



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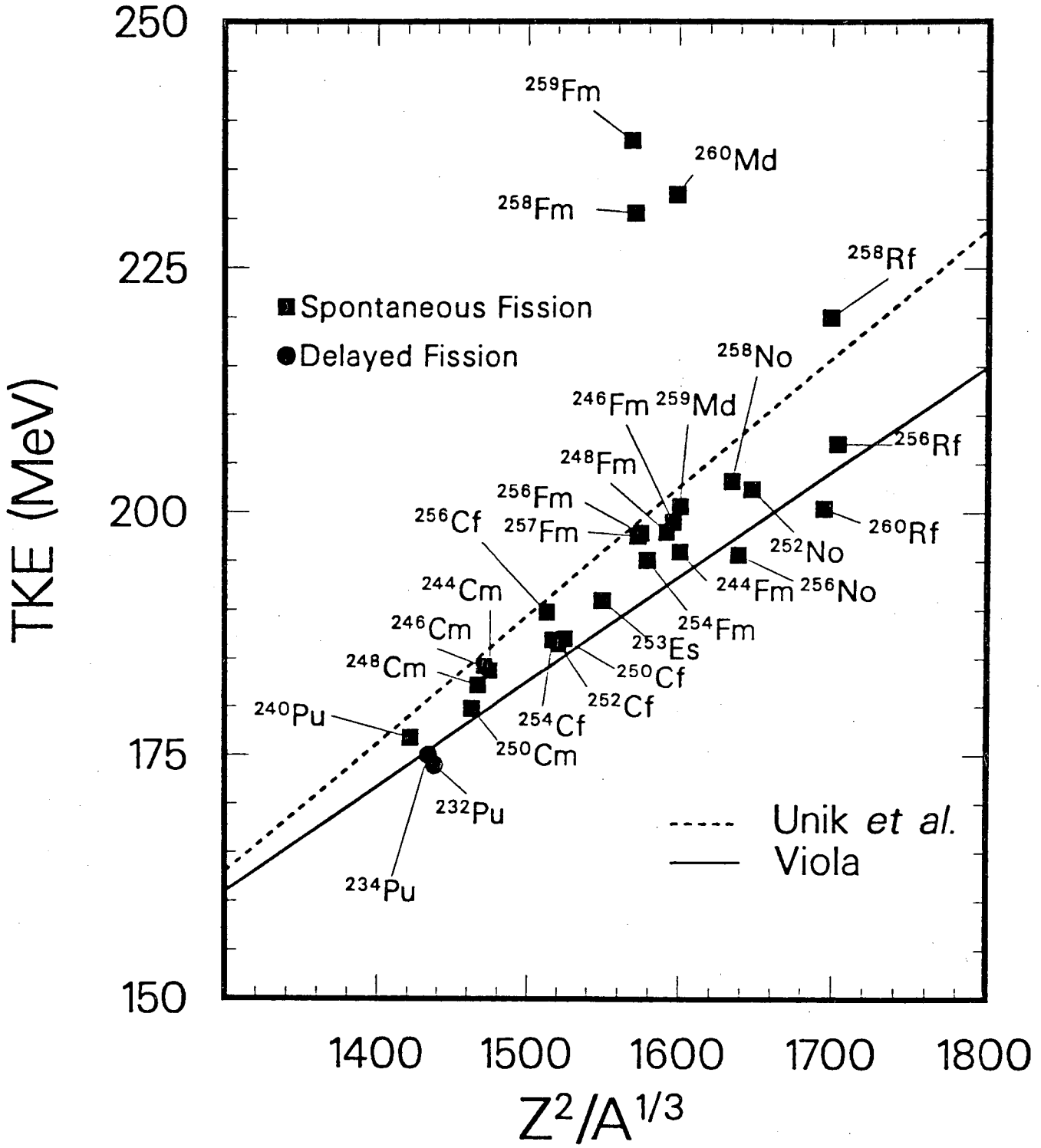
Hall & Hoffman - Fig. 3



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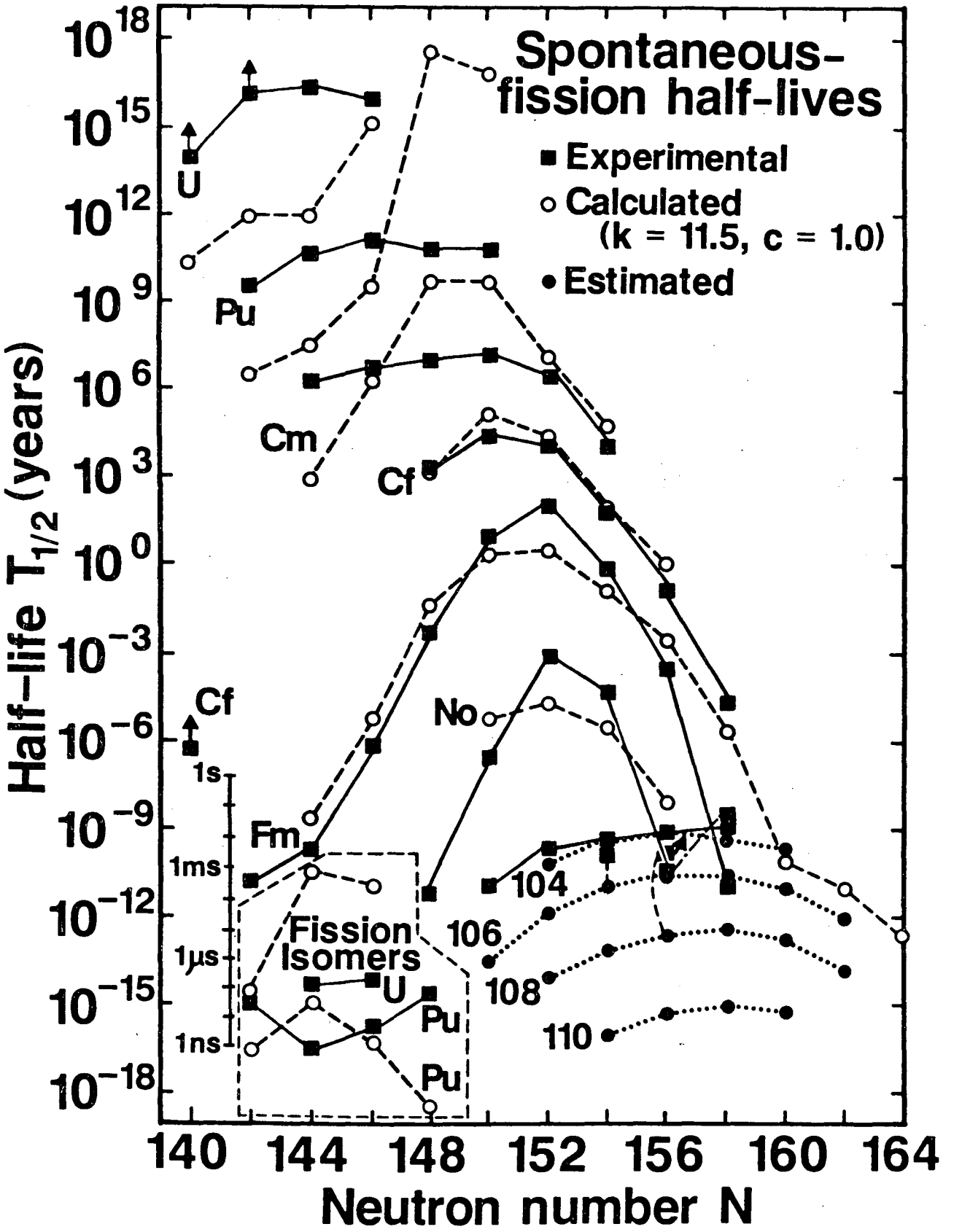
Hall & Hoffman - Fig. 4



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Hall & Hoffman - Fig. 5

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