Principles of Nuclear Fission

Gerhart Friedlander
Brookhaven National Laboratory, NY

Nuclear fission is the splitting of the nucleus of a heavy element such as uranium into two smaller nuclei. This important nuclear reaction which has had such profound impact on the world was discovered only a little over 50 years ago, in 1938, by two German chemists, Otto Hahn and Fritz Strassmann.

The discovery was totally unexpected and can be considered as a triumph of chemistry. All nuclear reactions known up to that time led to products within one or two atomic numbers of the target element. Thus, when Enrico Fermi and his collaborators in Rome irradiated uranium with neutrons, beginning in 1934, and found several series of beta–emitting radioactive species, it was natural for them to assume that these were isotopes of elements just beyond uranium. Subsequent work by the Rome group as well as by groups in Berlin and Paris confirmed and extended these results. Chemical tests showed that these products of uranium irradiation by neutrons did not behave like any of the known elements in the vicinity of uranium (uranium, protactinium, thorium, actinium, radium, etc.); i.e. in contrast to all these elements, they could be precipitated as sulfides in acid solution. This behavior, in fact, was what was expected for the "transuranium elements" of atomic numbers 93, 94, etc., which were then thought to be homologous with Re, Os, etc.

Thus, a sizeable literature grew up between 1934 and 1938 about several beta decay chains of "transuranium elements" designated as Eka–Re, Eka–Os, Eka–Ir.... as well as new isotopes of actinium and radium resulting from the uranium irradiations. Many of the results were very puzzling and also strained the concepts of nuclear physics — note that the different transuranium decay chains all were thought to have mass number 239, since they presumably all resulted from neutron capture in U-238! But the built–in prejudice against considering radically new concepts was so strong, especially among the physicists of the time, that nobody paid heed when the concept of fission was suggested by the German chemist Ida Noddack, the discoverer of the element rhenium. She had remarked already in a 1934 paper that Fermi should have compared the chemical properties of his new radioelements with those of <u>all</u> known elements, not only those in the vicinity of uranium, because, she wrote, "It is conceivable that in the bombardment of heavy nuclei with neutrons these nuclei disintegrate into several larger fragments which are indeed isotopes of known elements but not neighbors of the irradiated elements." So, here was the idea of fission; but Noddack, a chemist, was ignored by the physicists.

It took four years before the correct interpretation was established by painstaking chemical experiments of the kind that Noddack had suggested. The intervening history is fascinating but too long and complex to relate here. The decisive experiments of Hahn and Strassmann were intended to establish unambiguously the identity of several supposed radium isotopes by addition of barium carrier and fractional precipitation of barium bromide. It was well known that in this procedure radium is enriched in the initial precipitates. But much to the investigators' surprise, the supposed radium activities from the uranium irradiations remained uniformly distributed in all the barium fractions. To be absolutely sure of their result, they repeated the experiment with the addition of a known radium isotope, Ra–228, and found that it indeed behaved like radium, but the products of uranium bombardment behaved like barium. Despite their convincing chemical evidence, Hahn and Strassmann were very cautious in announcing their conclusion because "it contradicts all the experiences of nuclear physics to date," but their discovery was quickly confirmed by groups around the world.

As soon as the concept of fission was established, it became clear that all the so–called transuranium, actinium, and barium isotopes that had been produced by neutron irradiation of uranium were in fact isotopes of elements in the middle of the periodic table. (True transuranium elements were, of course, discovered subsequently, but that is a separate story.) Over 400 fission products have been identified, ranging from zinc (Z=30) to europium (Z=63). Their isolation and characterization in terms of mass numbers, half–lives, fission yields, radiations, and other properties has involved a vast amount of effort by radiochemists.

Why is fission of such great importance and interest? There are two key factors: 1) The energy release is huge — about 200 million electron volts (MeV) per fission or nearly 5×10^{15} kcal/mole! ; 2) The fact that about two neutrons are emitted per fission makes a chain reaction possible.

Both of these factors have to do with nuclear structure. The energy release comes about because nuclei near the middle of the periodic table are more tightly bound than those at either end (Fig. 1). Thus, when a uranium nucleus breaks up into two medium weight nuclei, the sum of the masses of the two products is less than the mass of the uranium nucleus, and the excess is released as energy (according to Einstein's $E=mc^2$), most of it as kinetic energy of the fragments. The emission of neutrons in the fission process occurs

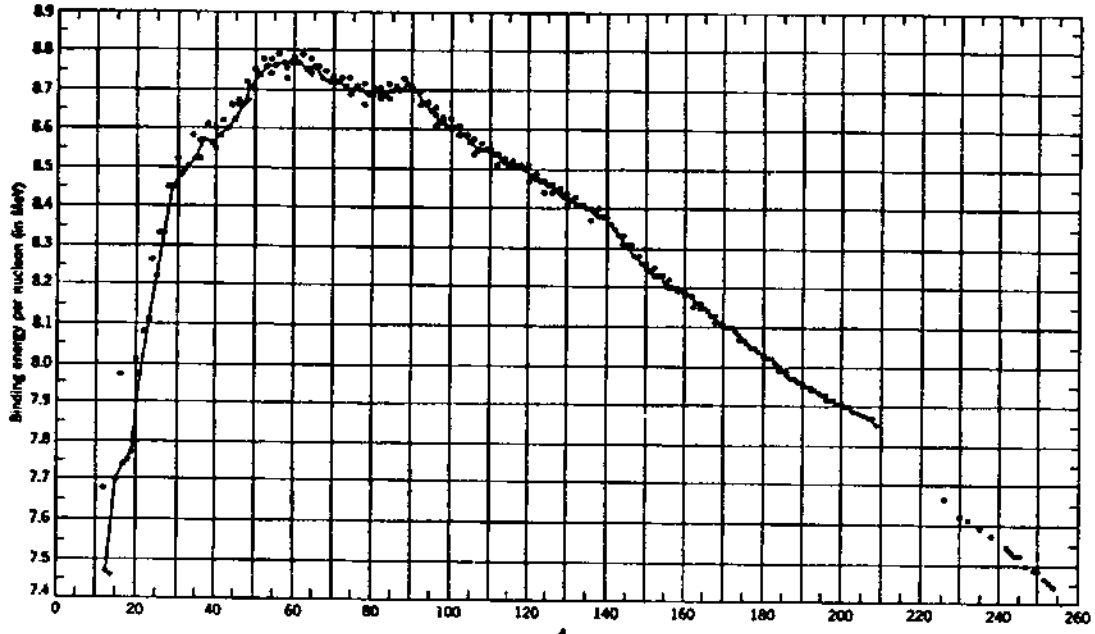

Fig.1 Average binding energy per nucleon as a function of mass number for stable nuclei. The line connects the odd–A points.

because stable nuclei of high Z have larger neutron/proton ratios than those of lower Z; a fact that is easily understood: the more protons that are packed into a nucleus, the more neutrons are needed to keep the protons apart; otherwise the Coulomb repulsion between the protons would make the nucleus unstable.

The large neutron/proton ratios of fissioning nuclei have another important consequence: even after the prompt emission of two or three neutrons in the fission process, the primary fission fragments have many more neutrons than the stable elements in their region of the periodic table. To illustrate: suppose a U–235 nucleus (which has 235–92=143 neutrons) captures a neutron, then fissions into Ba (Z=56) and Kr (Z=36) with the emission of two neutrons; there are then 142 neutrons to be distributed between the two fragments, yet the heaviest stable Ba isotope, Ba–138, has 82 neutrons, the heaviest stable Krypton, Kr–86 has 50. Thus, there are ten "extra" neutrons and the primary fission fragments are, therefore, far on the neutron–rich side of stability and highly unstable with respect to beta emission, e.g. Ba–144 (half–life 11.9 sec) and Kr–90 (half–life 32.3 sec).

The fact that the primary products are so far on the neutron–rich side of stability leads to chains of beta decays—the very chains some of which had erroneously been ascribed to chains of transuranium elements. Just to continue our example, Kr–90 decays to Rb–90 (15.3 sec) to Sr–90 (28.8 y) to Y–90 (64.1 h) to Zr–90 (stable). It is the relatively long–lived radioactive fission products (such as Sr–90) that give rise to the problems of waste management for the spent fuel elements from nuclear reactors.

One of the characteristic features of fission that emerged already from the early studies of fission products, largely by chemists, is the way the fission yields vary with product mass. This is shown in Figure 2 for thermal neutron fission of U–235. We see that the most probable split is into one heavy (A≈140) and one light (A≈95) fragment, whereas a symmetrical split is about 600 times less likely. The prevalence of

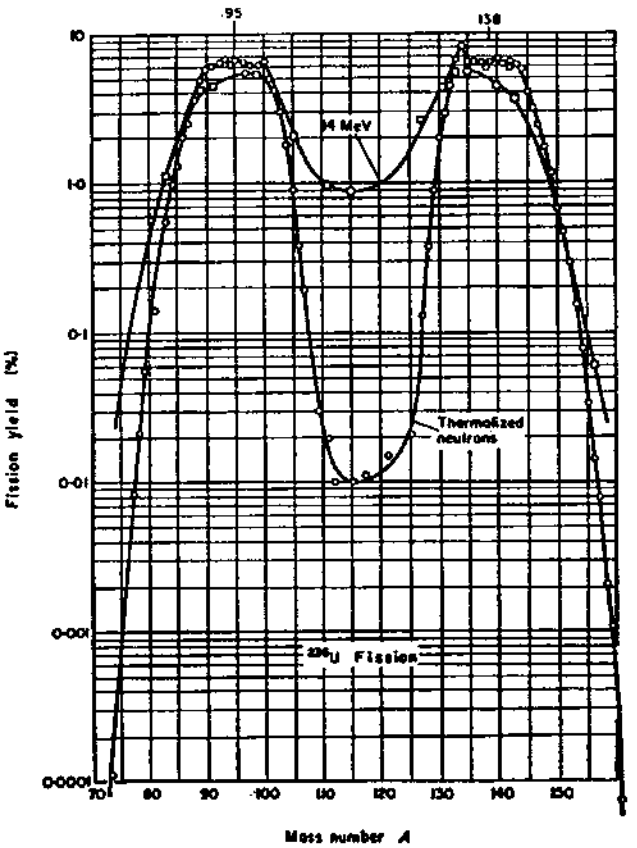

Fig. 2 Fission yield as a function of product mass number for fission of U–235 by thermal and 14–MeV neutrons.

asymmetric splits is characteristic of most fissioning systems, but with increasing energy of the bombarding particle symmetric fission becomes more and more probable. For example, in fission of U–235 with 14 MeV neutrons (prevalent in thermonuclear bombs) the ratio of asymmetric peaks to symmetric valley in the yield distribution is only about five (see Fig. 2). In some higher–Z systems, such as Fm (Z=100), symmetric fission actually prevails even at thermal neutron energies.

In addition to the distribution of mass among the products, the distributions of nuclear charge and kinetic energy are also important parameters of the fission process. We have already seen that (and why) the most probable products occur far on the neutron–rich side of stability. It turns out that the most probable split does not quite preserve the proton/neutron ratio of the fissioning nucleus; rather, the light fragment receives proportionately somewhat more of the total charge than its heavy partner. For a given A the dispersion of yields around that of the most probable product is a rather narrow Gaussian in Z (Fig. 3), with a width parameter σ of approximately 0.62 Z units (which means that one Z unit from the peak of the curve the yield is about 1/6 the maximum).

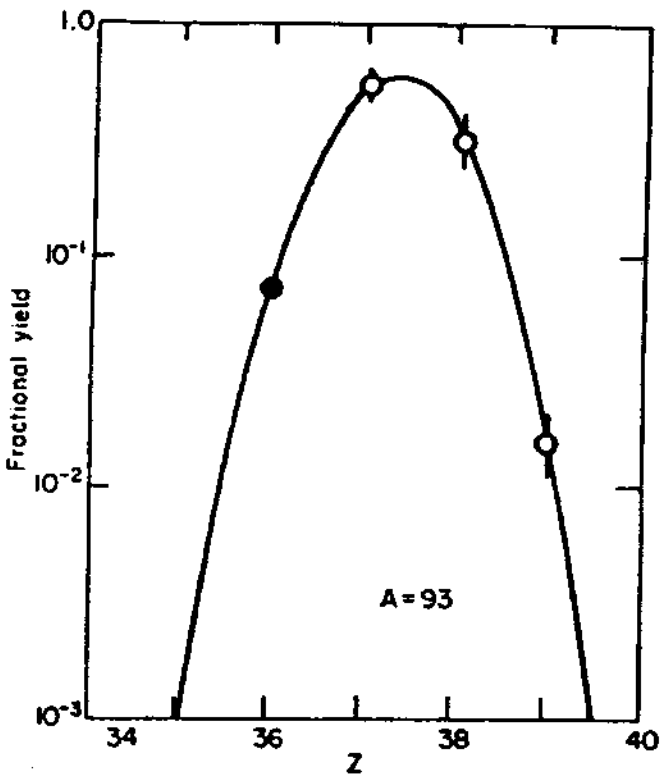

Fig. 3 Charge dispersion for fission products of mass number 93. The solid point represents the cumulative yield (including precursors) of 1.3 sec ^{93}Kr. The open circles are independent yields of ^{93}Rb (5.8 sec), ^{93}Sr (7.4 min), and ^{93}Y (10.2h).

As we already indicated, most of the energy released in fission goes into the kinetic energies of the fragments. For thermal neutron fission of U–235, the sum of the two kinetic energies is about 170 MeV on the average. However, the actual kinetic energy release varies with the mass split, being largest for a slightly asymmetric split ($M_h/M_l \approx 1.25$) and about 10–20% smaller for both symmetric and highly asymmetric mass divisions. Since the two fission fragments travel at 180°, with equal but opposite momenta, the kinetic energies for a given pair of fragments are inversely proportional to the masses. Thus, the mass distributions can be deduced from the kinetic energies of coincident pairs.

Since sustaining a chain reaction depends on the emission of more than one neutron per fission, much research has been centered on measurements of neutron emission. The average number of prompt neutrons per fission is 2.41 and 2.88, respectively, for the thermal neutron fission of U–235 and Pu–239. Most of the neutrons have been shown to originate from the fission fragments in flight, with the average number of neutrons per fragment being a rather strong function of the fragment mass (Fig. 4).

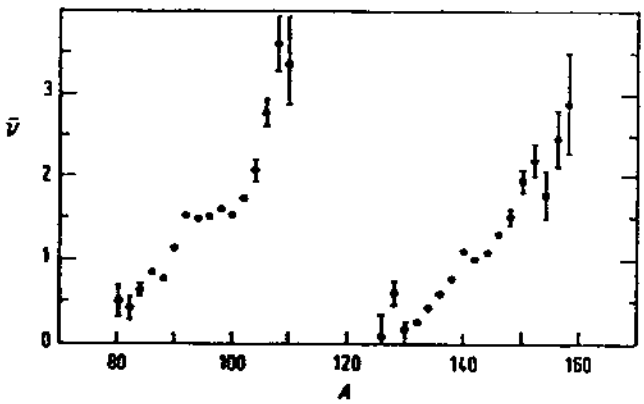

Fig. 4 Average number of prompt neutrons associated with fragments of different mass number formed in thermal neutron fission of U–235.

In addition to the prompt neutrons which are emitted in 10^{-14} sec, much smaller numbers are emitted with time delays of up to nearly a minute. These delayed neutrons have been shown to originate from certain fission products formed by beta decay in such highly excited states as to be unstable with respect to neutron emission. The preceding beta decay thus determines the half–life for neutron emission. Chemistry has been an important ingredient in identifying the specific beta–decay precursors of these neutron emitters. Over 60 of them are known and they play a crucial role in the control of nuclear reactors.

Although fission by thermal neutrons is surely the most important fission reaction — it is the basis of almost all working nuclear reactors — fission can also be induced by other projectiles including fast neutrons, charged particles such as protons and alpha particles, and photons. Fission can, in fact, even occur spontaneously. In the naturally occurring elements (U and Th), this process is so improbable that it is very hard to detect; but it becomes rapidly more probable with increasing Z, so that in the heaviest artificially

produced transuranium elements (Z>100) it is one of the dominant modes of radioactive decay.

The only naturally occurring uranium isotope that is fissionable by thermal neutrons is the rare isotope U–235 (natural abundance 0.72%). The other important species fissionable by thermal neutrons is Pu–239, produced by neutron capture in U–238 (to form U–239, half–life 23 min.), followed by two successive beta decays.

From a practical point of view (nuclear reactors, nuclear weapons) fission of only the heaviest elements (Z≥90) is of interest; but it should be noted that, with increasing energy of bombarding particles, it is possible to produce fission also in lighter nuclei, albeit with lesser probability. The probability of fission (as of all nuclear reactions) is expressed in terms of "cross-section". This, in naive terms, may be thought of as that cross–sectional area of the target nucleus which, if hit by the bombarding particle, leads to the specified reaction. Since nuclei have radii of 10^{-12} to 10^{-13} cm, nuclear cross-sections are typically less than 10^{-24} cm^2, and this has become the accepted unit of nuclear cross-section, named the barn (presumably because a cross-section as large as 10^{-24} cm^2 is "as big as a barn"). It is noteworthy that the thermal neutron cross-sections of U–235 and Pu–239 are 580 and 740 barns, respectively, whereas fast–neutron fission cross-sections never exceed the total geometric cross-sections of large nuclei, about two barns, and charged particle cross-sections are even smaller because of the Coulomb repulsion between nucleus and bombarding particle. The large values of some thermal neutron cross-sections have to do with the wave properties of thermal neutrons and can only be understood in terms of quantum mechanics.

The main purpose of this paper has been to give a brief introduction to the phenomenology of fission. The theoretical framework that has been developed to account for the phenomenon is beyond the scope of this discussion. However, a few words may be in order about the basic concept of a potential barrier against fission, which must be overcome by addition of energy, or penetrated by quantum–mechanical tunneling (as in spontaneous fission). In rough approximation, the height of the fission barrier is the difference between the Coulomb energy between the two fragments when they are just touching and the energy released in the fission process. The fact that in the region of uranium these two quantities happen to be nearly equal, each about 200 MeV, is responsible for the relatively low fission barriers in that region — between 5 and 6 MeV. It is this very delicate balance that causes U–235 to be fissionable with thermal neutrons whereas U–238 is not. The excited states of U–236 and U–239 formed by thermal neutron capture in U–235 and U–238, respectively, differ in excitation energy by only 1.7 MeV, but that is enough to account for one to be above, the other below, the top of the barrier.

For Further Reading

G. Herrmann, "Discovery and Confirmation of Fission", Nucl. Phys., **A502**, 141c–158c (1989)

E. P. Steinberg, "Radiochemistry of the Fission Products", J. Chem. Ed., **66**, 367–372 (1989)

E. K. Hyde, *The Nuclear Properties of the Heavy Elements, Vol. III – Fission Phenomena* , Prentice-Hall, New York, (1964)

R. Vandenbosch and J. R. Huizenga, *Nuclear Fission*, Academic Press, New York & London (1973)

J. E. Gindler and J. R. Huizenga, "Nuclear Fission" in *Nuclear Chemistry*, Vol. II, Leo Yaffe, Editor, Academic Press, New York (1968), 1–185