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Distributions of fission products from various low-energy fission reactions and the systematics of the odd-even fluctuations

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The dependence of the proton odd-even effect on the excitation energy of the fissioning nucleus and the mean kinetic energy of the fission fragments is examined in thermal neutron fission of ^{239}Pu and in "fast" neutron fission of ^{235}U, ^{238}U, and ^{232}Th. The proton odd-even effect observed in thermal neutron fission is found to decrease with increasing excitation energy and/or with increasing atomic number of the fissioning even Z nucleus. Possible factors responsible for the magnitude of the odd-even effect are discussed. An evaluation is made of a neutron odd-even effect, based on the observed mean proton odd-even effect, and consequently the mass distribution of fission fragments prior to neutron emission is calculated. A good fit is found between the calculated and the measured fragment mass yield curves and between calculated and measured neutron emission as a function of mass.

[NUCLEAR REACTIONS, FISSION ^{235}U(n,f), ^{239}Pu(n,f), ^{238}U(n,f), ^{232}Th(n,f), calculated odd-even effect; deduced systematics of the odd-even effect.]

I. INTRODUCTION

The distribution of the yields of fission products has not yet been determined with adequate accuracy other than for the fission of ^{235}U and ^{233}U induced by thermal neutrons.[1-3] Roughly, a Gaussian with a constant width of 0.56 ± 0.06 charge units does describe in a general way the fractional independent yields in any isobaric chain and in different fissioning systems.[4] Closer examination of even Z fissioning nuclides, however, revealed fluctuations about the Gaussian, so that the independent yields of fission products are higher than predicted by the calculations for those nuclides with an even number of protons and lower for those with an odd number of protons.[1-5] Some fluctuations have also been observed in the mass-yield distributions of fission fragments, i.e., before prompt neutron emission takes place. These observed fine structures follow a periodicity of five mass units and the peaks can be related to even Z_p values thus disclosing an odd-even (o-e) effect.[6,7] A quantitative approach to the o-e effect showed[1,2] that the average deviation from a "normal" distribution[4] was 22($\pm$7)% in the thermal fission yields of both ^{235}U and ^{233}U.

Since the publication of the above o-e effect,[1,2] it has been used in several compilations of fission product yields for various fissioning nuclei and neutron energies ranging from thermal to 14 MeV.[8-10] This was done without any experimental evidence for the dependence of the o-e effect on the energy and fissioning nucleus. Careful examination of published fission yields of uranium and plutonium isotopes, as well as recently determined fission yields in the fast neutron fission of ^{232}Th,[11] now facilitates a more rigorous treatment of the systematic behavior of the o-e effect in the distribution of nuclides in fission, and the dependence of this effect on the fissionable nuclide in question and on the excitation energy. Hence, more accurate calculations of fission yields may be obtained.

The observed o-e effect suggests that a partial preservation of proton pairs during fission takes place despite the relatively high energy release accompanying this process. This indicates that there must be a delicate energy balance among the various degrees of freedom, even at the rapid transition between the saddle and scission configurations of the fissioning system. Analogously, the above reasoning should also apply to the preservation of neutron pairs.

An examination of the yields of fission products reveals only a relatively small neutron pairing effect. An average o-e effect of ~8% was measured in the isotonic fission yields as a function of neutron number in the heavy mass peak,[2] and a similar effect in the heavier side of the light mass peak has been more recently observed in on-line mass spectrometer measurements.[3] This effect may be interpreted by primary neutron pair splitting and/or as a result of subsequent neutron evaporation; these points will be discussed later. Nevertheless, one should expect a neutron odd-even effect in the pair breaking analogous to the one observed for protons, because the pairing energies in both cases are roughly of the same magnitude. Consequently, a method can be proposed for the calculation of independent yields of individual fission fragments, based on measured yields of the elements and on a Gaussian of constant width representing the isobaric dispersion[12] on which odd-even fluctuations for neutrons and protons are superimposed.

II. ANALYSIS OF EXPERIMENTAL RESULTS

The odd-even effect observed in the thermal neutron fission of ^{235}U and ^{233}U is based on substantial experimental evidence.[1,2] Its magnitude can be obtained by comparing the yields of elements with odd and even numbers of protons, as well as from the deviations of the measured yields of individual isotopes from the normal distribution, as suggested by Wahl.[4,12] In other fissioning systems it is more difficult to derive an accurate value for the odd-even effect, due to the paucity of experimental data and the uncertainties concerning the normal distribution. Though the width of the Gaussian representing the dispersion of the isobaric yields fluctuates about a constant value within a wide range of excitation energies and for various fissioning nuclei,[13] the most probable charge Z_p is subject to changes depending on the mass charge and excitation of the fissioning nucleus.[14] In general, Z_p values in any fissioning nucleus may be calculated relative to Z_p^{ref} values of a reference reaction, usually the fission of ^{235}U induced by thermal neutrons. Thus we obtain

$$Z_p = Z_p^{ref} + \Delta Z_p . \qquad (1)$$

The ΔZ_p values are calculated from Eq. (2) using semiempirical constants A, B, and C:

$$\Delta Z_p = A(Z_F - Z^{ref}) + B(A_F - A^{ref}) + C(\nu_T - \nu_T^{ref}) . \qquad (2)$$

where Z_F, A_F, and ν_T are the nuclear charge, mass, and total number of neutrons emitted in fission, respectively.

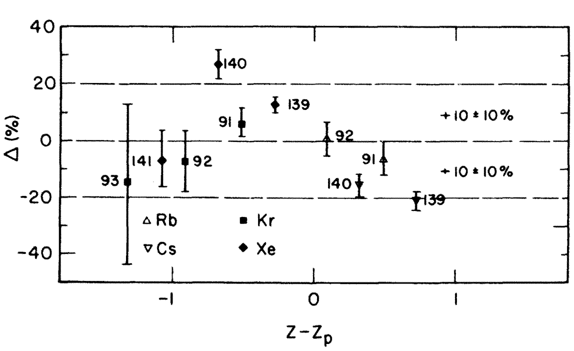

FIG. 1. Deviations from normal distribution in the yields of fission products in fast neutron fission of ^{235}U. The broken lines represent one standard deviation about the mean value.

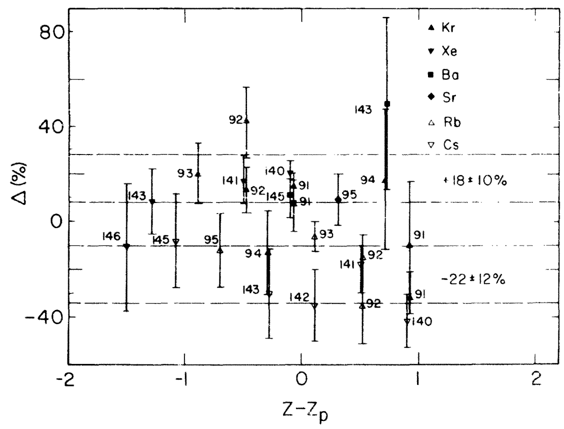

FIG. 2. Like Fig. 1, for fast neutron fission of ^{238}U.

In this study, the odd-even systematics in the thermal neutron fission of ^{239}Pu and in the fast neutron (fission spectrum) fission of ^{235}U, ^{238}U, and ^{232}Th were investigated. In each case the experimental yields were compared with normal Gaussian distributions based on Z_p values calculated by Nethaway and Barton[15] and a constant width of 0.56 ± 0.06 charge units.[4]

In the calculation of the Z_p values[15] according to Eq. (2), different semiempirical constants were applied for the light and the heavy mass peaks and the resulting Gaussians representing the normal distribution gave better fits with measurements than normal distributions based on Z_p values calculated from a single set of semiempirical constants.[9,16,17] The above finding is consistent with the fact that in low-energy fission the mean mass of the heavy mass peak remains constant around mass 140–142 irrespective of the fissioning nuclide, while the mean mass of the light mass peak shifts towards higher values in heavier fissioning nuclei. The deviations of the experimental values of the fractional independent yields from the normal ones are represented in Figs. 1–4 for fast neutron fission of ^{235}U, ^{238}U, and ^{232}Th and for thermal neutron fission of ^{239}Pu. The experimental data were taken from the work of Wolfsberg[18] for fast neutron fission of ^{235}U, from Wolfsberg,[18] Balestrini and Forman[19] for fast neutron fission of ^{238}U, and from Izak-Biran and Amiel[11] for the fast fission of ^{232}Th. The fission yields in thermal neutron fission of ^{239}Pu are better known, and Fig. 4 is based on the works of Qaim and Denschlag,[20] Wolfsberg,[21] Balestrini and Forman,[22] Okasaki, Walker, and Bigham[23] and Hawkings, Edwards, and Olmsted.[24]

Although, as emphasized above, the experimental data are still incomplete, an average odd-even effect may be derived for each case and a general

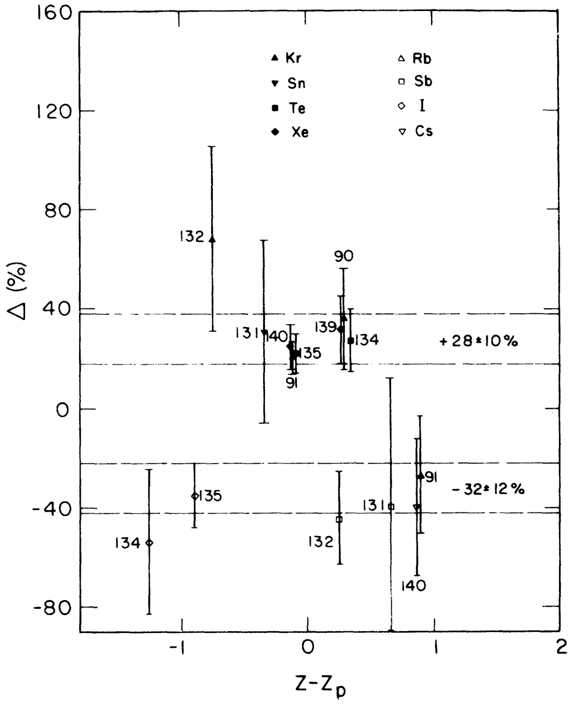

FIG. 3. Like Fig. 1, for fast neutron fission of ^{232}Th.

trend in the odd-even systematics may be observed. (Here, the o-e effect is defined as the difference between the experimental and calculated normal independent fractional yields, divided by the calculated normal independent fractional yield.)

The trend of the o-e effect in the various fission reactions may be correlated with two major parameters, viz., the excitation energy at the saddle above the fission barrier (Fig. 5) and the atomic number of the fissioning nuclide (Fig. 6). From Fig. 5 it may be deduced that the o-e effect

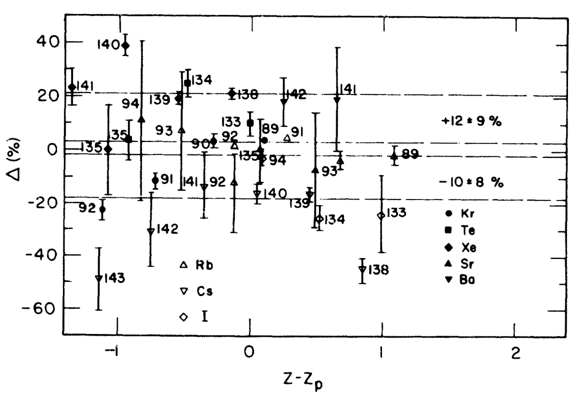

FIG. 4. Like Fig. 1 for thermal neutron fission of ^{239}Pu.

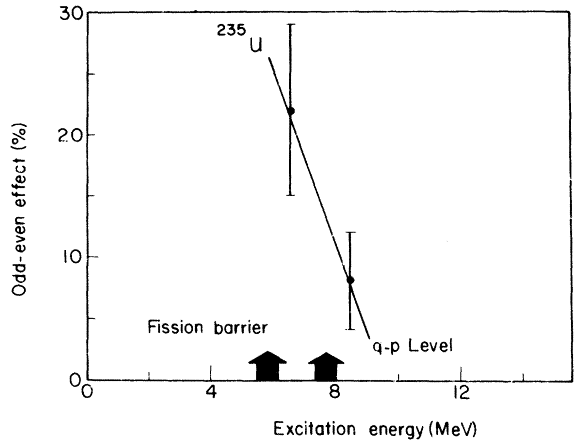

FIG. 5. Dependence of the odd-even effect on the excitation energy at the saddle in the fission of ^{235}U, q-p level—the particle-hole level above the saddle.

should disappear at high excitation energies, unless the emission of prescission particles brings about substantial deexcitation of the fissioning nucleus. Likewise, in high Z nuclei (>94) the effect is remarkedly reduced and seems to disappear.

The o-e effect in various fissioning nuclides, based on experimental fission yields, is summarized in Table I, which also gives information

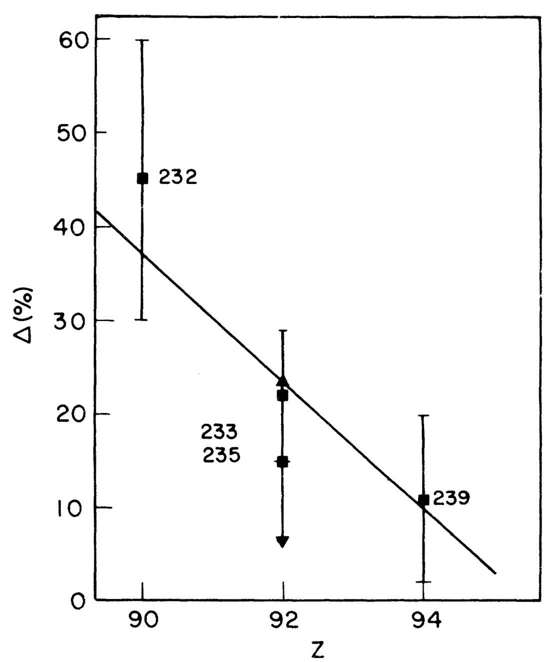

FIG. 6. Dependence of the odd-even effect on the atomic number of the fissioning nucleus (thermal fission).

TABLE I. The odd-even effect based on experimental fission yields.

Isotope	Mean neutron energy (MeV)	Fission barrier (MeV)	Excitation energy (MeV)	Total kinetic energy (MeV)[a]	Odd-even effect (%)
^{233}U	0	6.0 + 0.2 [b]	6.9	167.49	22 ± 7 [c]
^{235}U	0	5.9 ± 0.2 [b]	6.6	167.91	22 ± 7 [c]
^{235}U	1.9	5.9 ± 0.2 [b]	8.5		10 ± 10 [d]
^{238}U	1.9	6.2 ± 0.2 [b]	6.7	165.89	20 ± 11 [d]
^{239}Pu	0	5.4 [e]	6.8	173.97	11 ± 9 [d]
^{232}Th	1.9	6.4 ± 0.2 [b]	6.7	160.02	38 ± 13 [f]

[a] Unik and Gindler, Ref. 44.
[b] Vandenbosch and Huizenga, Ref. 27.
[c] Amiel and Feldstein, Ref. 2.
[d] This work.
[e] Tsang and Wilhelmy, Ref. 26.
[f] Izak-Biran, Ref. 11. The measured o-e effect of 30(±12)% was corrected for the particle-hole excitation at the saddle, induced by the energetic part of the neutron spectrum.

concerning the heights of the fission barriers, excitation energies, and total kinetic energies of the fission fragments.

III. DISCUSSION

A. Excitation energy and particle-hole states

The odd-even effect found in low-energy fission of even-even uranium and thorium isotopes indicates that at least some of the primary proton pairs are preserved, even when the nucleus fissions. Consequently, energy in excess of that needed for ground state deformation is required to break the existing proton pairs in the deformed nucleus. The proton pairing energy at the saddle configuration is about 1.7 MeV (Ref. 25) and if the energy is available, the particle-hole excited states may be filled by single nucleons.

At scission, the distribution of nucleons in the particle-hole states among the fission fragments is assumed to be random. Therefore, if those states are populated, there is an equal probability for the formation of odd or even fragments. The extent of particle-hole excitation is expected to be proportional to the available energy at the saddle. This reasoning is consistent with the experimentally found drop in the odd-even effect in the fast neutron fission of ^{235}U,[2] as compared with thermal fission of ^{235}U (Fig. 5). The fission barriers and particle-hole states in ^{234}U, ^{236}U, ^{238}U, ^{240}Pu, and ^{233}Th are shown in Fig. 7. (The heights of the fission barriers were taken from Tsang and Wilhelmy[26] and Vandenbosch and Huizenga.[27])

As may be seen from Fig. 7, the absorption of a thermal neutron by ^{235}U, ^{233}U, or ^{239}Pu, or a fast neutron (mean energy of 1.9 MeV) by ^{238}U or ^{232}Th, supplies the fissioning nuclide with sufficient energy to overcome the fission barrier, but the residual excitation is short of the amount required for breaking proton pairs and filling the particle-hole states. Nevertheless, in all the cases mentioned above, the formation of primary odd-Z fission fragments was observed experimentally. These observations imply that the "pair breaking" process is more complex and may take place between the saddle and the scission whenever sufficient energy is available. Since the experimental mean o-e effect in low-energy fission is much smaller than could be expected (from the externally supplied excitation energy), excitation during the descent from saddle to scission involves a process of energy conversion responsible for the proton pair breaking. A possible source of additional energy is the kinetic energy of the fragments before scission. This energy may be partly converted into single particle excitation, which indicates that the mass flow of the deformed nucleus is not completely adiabatic.

On the whole the energy transfer involved in the particle-hole excitation is about 2 MeV, while the total kinetic energies in the nuclei in question range from 160 to 180 MeV (Table I). More direct experimental evidence in support of the above conclusion concerning the partial viscosity of the mass flow can be found in the fluctuations in the kinetic energies of fragments from spontaneous fission of ^{252}Cf (Ref. 28) and thermal neutron fission of ^{229}Th (Ref. 7) measured as a function of the proton number. Other evidence is the emission of charged particles or neutrons from the "neck" of the deformed fissioning nucleus prior to scission.[29]

On the other hand, pronounced fluctuations in the mass yield curves, consistent with the o-e

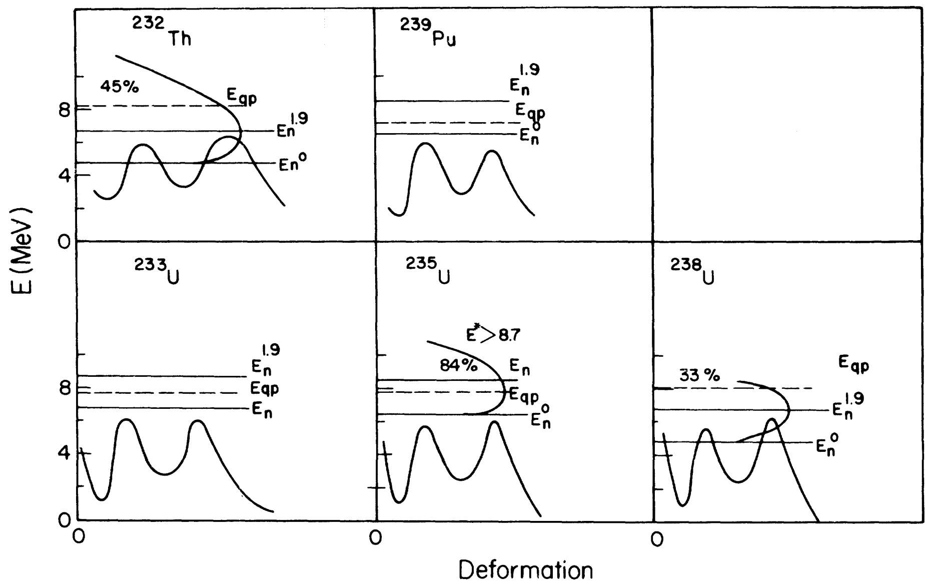

FIG. 7. Fission barriers and particle-hole states in ^{234}U, ^{236}U, ^{239}U, ^{240}Pu, and ^{233}Th. E_{qp}—energy of the particle-hole state, E_n—energy induced by the absorbed neutron, E_n^0—thermal neutron, $E_n^{1.9}$—mean neutron energy 1.9 MeV. The heights of the fission barriers were taken from Tsang and Wilhelmy (Ref. 26) and Vandenbosch and Huizenga (Ref. 27).

proton configuration, were observed in selected fission events with kinetic energies higher than average.[3,30,31] Even in cases where the average o-e effect is insignificant, like the thermal neutron induced fission of ^{239}Pu [o-e effect of 11(±9)%, Table I], a substantially marked fine structure was found in the mass curve of fission fragments at kinetic energies higher by 10 MeV than average.[6]

In the thermal neutron fission of ^{235}U, where the average o-e effect is 22(±7)%, the fluctuations of the mass yield curves at discrete kinetic energies become more pronounced with the increase in the kinetic energy of fragment pairs.[30,31] What is interesting is that some structure is still observed at kinetic energies below the average, where the expected excitation energy is high.[31] This may indicate that the decrease in the kinetic energy is divided between the excitation at the saddle and deformation of the fission fragments, the latter competes with proton pair breaking.

1. Dependence of the o-e effect on the excitation energy at the saddle

The odd-even effect in fast neutron fission relative to that in thermal fission may be roughly calculated if one assumes the following:
(a) The number of particle-hole excitations at the saddle is directly proportional to the number of neutrons which have sufficient energy to induce that excitation.
(b) The percent of particle-hole excitations during the descent from saddle to scission relative to the number of ground-state proton pairs is the same as in thermal neutron fission.
As an example, the fast neutron fission of ^{235}U is considered. In this case, absorption of a neutron with kinetic energy of 1.9 MeV results in a total excitation of 8.5 MeV, while the fission barrier is only 5.9±0.2 MeV; the difference of 2.6 MeV is somewhat higher than the proton pairing energy.[25]

The energy spectrum of fission neutrons is such that about 34% have energy higher than 1.9 MeV,[32] and therefore are capable of bringing about particle-hole excitation. Of the residual 66%, only 78% are excited to particle-hole levels [based on an original 22(±7)% o-e effect], and therefore the calculated o-e effect in this case is 14.5(±10)%, while the one based on measured yields is 10(±10)% (Fig. 1).

2. Dependence of the o-e effect on the atomic number of the fissioning nucleus

The o-e effect as a function of the atomic number of even Z nuclei in the range $90 \leq Z \leq 94$ may be approximately represented by a straight line with a negative slope, i.e., the o-e effect decreases with increasing atomic number (Fig. 6). Actually, the o-e effect may be represented as a function of any of several Z-dependent parameters, such as kinetic energy, fissility parameter, or level density, and it cannot be stated with certainty which is the determining factor in the systematics of the o-e effect.

Correlation with the mean kinetic energy indicates that the particle-hole excitation increases (o-e effect decreases) with increase in energy which is consistent with the nature of viscous flow, where the friction is proportional to the kinetic energy. On the other hand, measurements of fission yields of the same fissioning nuclide but at selected kinetic energies showed that the o-e effect increases at high kinetic energies, provided that the overall excitation is low. Consequently, correlation of the o-e effect with the kinetic energy of the fragments is not a simple function which may be easily calculated.

Björnholm[33] examined the particle-hole excitation during the descent from saddle to scission as a function of the fissility parameter $a = Z^2/A$, which determines the shape of the fission barrier. He related the particle-hole excitation to the fine structure found in the mass yield curves of fission fragments in various fissioning nuclei. In general, he found a decrease in the fine structure with the increase of the fissility parameter, with the exception of the spontaneous fission of ^{248}Cm, where the structure is more pronounced than in the case of thermal neutron fission of ^{239}Pu. To explain this, an especially wide fission barrier was proposed, so that the descent from saddle to scission starts at a considerably elongated shape and the deformation energy decreases the available excitation energy required for pair breaking. Another explanation, based on the same fission barrier model, is that the number of single nucleon level crossings decreases[34] with increasing deformation and therefore more proton pairs may be preserved.

The decrease in the o-e effect at higher atomic numbers may also be explained by increasing density of the particle-hole levels at the descent from saddle to scission with increasing Z, which facilitates more level crossings and subsequent proton pair breaking. At any rate, all explanations of the systematics of the o-e effect are closely related to the atomic number of the fissioning nucleus, and therefore the presentation of the o-e effect as a linear function of Z in the range $90 \leq Z \leq 94$ (Fig. 6) is coherent with each of the above explanations. Extrapolation of the o-e effect to higher even Z elements does not suggest any o-e effect in the fission of curium isotopes, although the unseparated mass yield curves from spontaneous fission of the isotopes ^{246}Cm and ^{248}Cm exhibit a pronounced fine structure in one report[7] while in another work on the distribution of fission yields in the spontaneous fission of ^{246}Cm, the structure of the mass yield curve is insignificant.[35]

Correlation of the o-e effect with the width of the fission barrier was not considered in our work, since the data concerning this parameter differ considerably in the various calculations.[36-38]

B. Neutron odd-even effect

The o-e effect in the distribution of elemental yields in fission is due to the proton pairing energy, which is about 1.7 MeV (Ref. 25) in a deformed nucleus in the saddle configuration. In the ground state configuration, the neutron pairing energy is very close to[39] or even as much as 20% higher than the proton pairing energy.[25] This can lead one to assume that proton and neutron pairs should be treated in a similar way in deformed nuclei as well. If this assumption is valid, then an o-e effect in neutrons at least of the same magnitude as that for protons may be expected. This primary effect in fission fragment formation cannot be verified by direct measurements, due to the masking effect of prompt neutron emission (approximately 10^{-14} sec). Therefore, all the measured mass distributions are shifted with respect to the pre-neutron-emission fragment distribution, and any initial fine structure due to the neutron pairing is obliterated. The observed o-e effect of ±8% in the isotonic yields[1-3] is dominated by neutron emission, which washes out most of the primary effect.

C. Tentative model for the distribution of fission fragments

Even though the neutron o-e effect in the distribution of fission fragments cannot be measured directly, a semiempirical model for the distribution of fission fragments (prior to neutron emission) may be proposed, in which an o-e effect, equal for both protons and neutrons, is assumed. The model is based on the following data and assumptions:

(1) Empirical yields of the elements (^{236}U data were taken from Refs. 1 and 2).

(2) A Gaussian distribution of constant width of the isobaric, pre-neutron-emission chains ($\sigma = 0.40$ charge units for ^{236}U) taken from x-ray measurements of fission fragments.[40]

(3) An o-e correction factor multiplying a normal Gaussian distribution, as also presented by Reisdorf, Unik, and Glendenin.[30]

(4) A_p, the most probable mass of the isotopic distribution, taken from Reisdorf *et al.*[40]

(5) The isotopic distribution assumed to be Gaussian with a constant width $\sigma_{\text{isotopic}} = \sigma_{\text{isobaric}} A_F/Z_F$, with a superimposed neutron o-e effect. (Here A_F and Z_F are the mass and atomic number of the fissioning nucleus.)

Thus we obtain for a fission fragment of mass A

and atomic number Z

$$ {}^{A}_{Z}y = \frac{{}_{Z}Y(1\pm a)}{(\pi c)^{1/2}} \int_{A-1/2}^{A+1/2} e^{-(n-A_p)^2/c}\, dn\,, \qquad (3) $$

where ${}^{A}_{Z}y$ is the fission yield of fission fragment of mass A and atomic number Z, ${}_{Z}Y$ is the fission yield of element of atomic number Z (includes the proton o-e effect), a is the o-e neutron effect (22% for the thermal neutron fission of ^{235}U), and c is a constant dependent on the width of the Gaussian σ, $c=(2\sigma^2+1/12)$.

The validity of the above model may be checked by comparison with experimental measurements. Such a comparison is made in Fig. 8 (mass distribution) and Fig. 9 (prompt neutron emission). The calculated mass distributions exhibit pronounced fluctuations which are considerably moderated after correction for the experimental mass resolution of $\sigma=1.5$ mass units,[30] and good agreement with the experimental results is obtained.

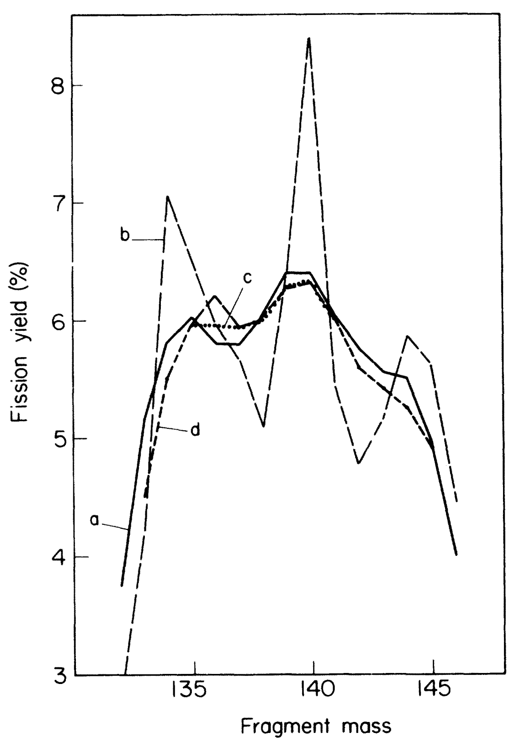

FIG. 8. The distribution of yields of fission fragments in the thermal neutron fission of ^{235}U. a—experimental curve, Unik *et al.* (Ref. 7), b—calculated curve, assuming a Gaussian isotopic distribution with $\sigma=1.03$ amu and a 10% odd-even neutron effect. Uncorrected for resolution. c—like b, but 22% odd-even effect and corrected for resolution with $\sigma_D=1.5$ amu. d—like c, but 10% odd-even effect.

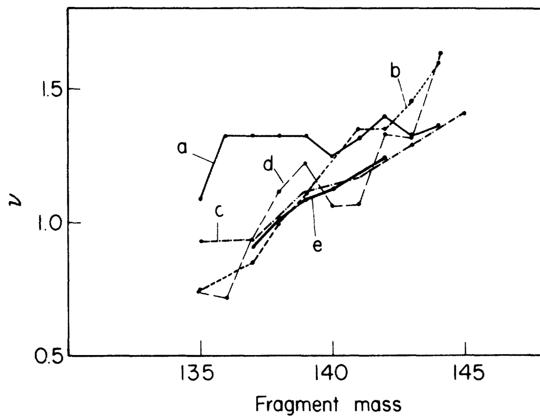

FIG. 9. Prompt neutron emission in the heavy mass peak, in thermal neutron fission of ^{235}U. a—Terrel (Ref. 41), b—Apalin *et al.* (Ref. 42), c—Boldeman, Musgrove, and Walsh (Ref. 43), d—this work, uncorrected for resolution, and e—this work, corrected for resolution.

The considerable spread due to the experimental resolution obliterates any fine structure and the comparison is insensitive to the magnitude of the o-e effect (Fig. 8).

The prompt neutron emission function was calculated from the difference between the calculated pre-neutron yields and the experimental post-neutron yields by a method similar to that of Terrel[41] and the results, after correction for mass resolution, fall well within the range of the experimental results (Fig. 9). Here again, due to the experimentally reported mass resolutions, the magnitude of the assumed effect cannot be clearly determined.

The calculations of the yields of fission fragments and neutron emission, discussed above (Figs. 8 and 9), are based on considerable experimental data.[40] However, for many fissioning systems these data are unavailable. In those cases where only the mass yields (prior to neutron emission) are known, the calculations are based on the normal distribution[4] and calculated Z_p values.[15] Hence, the elemental yields may be summed up and corrected for the o-e effect according to o-e systematics. Once the elemental yields are known, the yields of fission fragments may be calculated according to Eq. (3).

In conclusion the proposed method permits the calculation of the yields of fission fragments, based on the o-e effect for both protons and neutrons. This method is justified by energy considerations and in the case of thermal neutron fission of ^{235}U a good fit was obtained between the calculated and measured yields.

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