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Semiempirical Correlation of Fission Yield and Kinetic Energy Distributions

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IN the Wigner-Eisenbud formalism the configuration space occupied by a disintegrating nuclear system is divided by a boundary into an internal region in which nuclear interactions dominate and an external region in which the separated pairs of fragments interact only through their mutual Coulomb fields. The total probability of obtaining disintegration through some specific channel is equal to the internal reduced width for the channel multiplied by external factors which are the wave number associated with the fragments and the Coulomb barrier penetrability evaluated at the boundary.

Fong[1,2] and Newton[3] have attempted to calculate the yields and kinetic energy distributions of fission fragments using this formalism. The basis of their approach is the assumption that the internal reduced widths are the same, on the average, for all fission channels. The problem then becomes one of computing the external probabilities for the different channels and summing over all channels which form specific pairs of fission fragments. Newton[3] has reached the conclusion that this method fails when proper shell-dependent nuclear level densities are used in the summation.

The writer[4] has recently obtained an improved level density formula in which the dependence on closed shells has been incorporated by a procedure which relates the level density parameters to the ground-state masses of nuclei. The procedure reproduces 82 observed level spacings with a root mean square error factor of 1.74. This revised formula has been used in a fresh calculation based on the Fong-Newton procedures.

Following Newton,[3] we write the yield of a specific pair of fission fragments as

$$Y(Z_L,A_L,Z_H,A_H)=f_i\int_0^{E_T}dE_K\int_0^{E_T-E_K}dE_L\frac{\mu^2E_K}{F^2+G^2}\times\omega_L(E_L)\omega_H(E_T-E_K-E_L),\quad(1)$$

where Z_L and A_L are the charge and mass numbers of the light fragment, Z_H and A_H those of the heavy fragment, f_i is an internal probability which is assumed to be constant, E_T is the total energy release of the fission mode minus the pairing energy of the fragments, E_K is the kinetic energy of the fragments, E_L is the excitation energy of the light fragment, and ω_L and ω_H are the level densities of the two fragments at the indicated excitation energies. μ is the reduced mass of the fragments given by $\mu=A_LA_HM_N/(A_L+A_H)$, where M_N is the mass of a nucleon. The barrier penetrability is given by

$$\frac{1}{F^2+G^2}=0.5466\left(\frac{B}{E_K}\right)^{5/3}\frac{(Rk)^{-\frac{1}{3}}}{\bar{K}(x)},\quad E_K>B$$

$$=0.5466\left(\frac{B}{E_K}\right)^{5/3}\frac{(Rk)^{-\frac{1}{3}}}{K(x)}e^{-2x},\quad E_K\leq B.$$

Here

$$B=Z_LZ_He^2/R,$$

$$x=\frac{2}{3}\frac{(Rk)}{E_KB^{\frac{1}{2}}}(|B-E_K|)^{\frac{3}{2}},$$

$$K(x)=e^{-2x}x^{\frac{2}{3}}[I_{\frac{1}{3}}^2(x)+I_{\frac{1}{3}}(x)I_{-\frac{1}{3}}(x)+I_{-\frac{1}{3}}^2(x)],$$

$$\bar{K}(x)=x^{\frac{2}{3}}[J_{\frac{1}{3}}^2(x)-J_{\frac{1}{3}}(x)J_{-\frac{1}{3}}(x)+J_{-\frac{1}{3}}^2(x)],$$

e is the electronic charge, R is the radius of the boundary between the internal and external regions of configuration space, and J and I are Bessel functions.

Equation (1) is a summation of all the ways in which the effective energy release E_T can be divided between the kinetic and excitation energies of the fragments and of the ways in which the excitation energy can be split between the fragments. It should be noted that no summation over angular momentum combinations has been made. This summation was omitted because the barrier penetration probability decreases only very slowly as the relative orbital angular momentum of the fission fragments increases, and it was therefore felt that the summations over all these momenta and of the resulting combinations of spins of the excited states of the fragments would result in multiplying the yields of Eq. (1) by a factor which is nearly the same for all fission modes.

The effective energy release E_T has been defined as the total energy release minus the pairing energies of the fragments because the level density formula[4] is dependent on effective excitation energies which exclude pairing energies.

The energy released in all the significant fission modes in the thermal neutron fission of U^{235} was calculated from the writer's revised semiempirical atomic mass formula,[5] and pairing energies[4] were subtracted to give

[1] P. Fong, Phys. Rev. **89**, 332 (1953).
[2] P. Fong, Phys. Rev. **102**, 434 (1956).
[3] T. D. Newton, Chalk River report CRP-642-A, p. 307.
[4] A. G. W. Cameron, Can. J. Phys. (to be published).

[5] A. G. W. Cameron, Can. J. Phys. **35**, 1021 (1957).

the effective energy releases E_T. The integrals in Eq. (1) were performed numerically for several values of R in order to give predictions of fission yields. It was found that the resulting yields bore little resemblance to the yields determined experimentally. The relative shape of the mass yield curve which was calculated was not a very sensitive function of the assumed value of R. The yields were sharply peaked at a mass ratio of 1.34 and there was only a very shallow valley. This confirms Newton's earlier conclusion[3] that the method fails to reproduce the mass yield curve.

Since it had become desirable to obtain predictions of independent fission yields in connection with the writer's work in nuclear astrophysics,[6] an attempt was made to construct a model out of the Wigner-Eisenbud formalism by specifying the radius of separation of the fragments to be a function of the fission channels. It became apparent that in order to give a simultaneous fit to the experimentally determined yields and kinetic energy distributions, the radius must depend on the fission modes in two ways. Firstly, the radius of separation characteristic of symmetric fission must be significantly smaller than that characteristic of asymmetric fission. Secondly, the radius of separation of the fragments must increase when more of the energy release in fission is stored as excitation energy of the fragments. This lowers the Coulomb barrier and increases the width of the kinetic energy distribution.

The radius of separation was therefore written in the form,

$$R = R_0 + \alpha (E_T - E_K)^{3/2}. \tag{2}$$

The first term, R_0, depends on the mass ratio of the fission fragments. The second term is attributed to the "fringe expansion" of the nucleus. As the excitation energy of the nucleus increases, some of the nucleons in low-lying orbits are raised to orbits considerably above the Fermi level of the nucleon gas. The nucleon wave functions then extend to larger distances in configuration space than before, and the nuclear surface becomes more diffuse.

Fraser and Milton[7] have recently determined the half-width of the kinetic energy distribution of fission fragments from the most probable modes in the spontaneous fission of Cf^{252} to be 17 Mev. Milton[8] estimates that the existing measurements of the similar quantity in the thermal fission of U^{235} are most consistent with a half-width of about 14.5 Mev. These values can be fitted with the form of the second term given in Eq. (2) and with

$$\alpha = 0.0055 \text{ fermi Mev}^{-3/2}, \tag{3}$$

where 1 fermi $= 10^{-13}$ cm. It should be noted that unless the second term depends on the excitation energy raised to a power greater than unity it is not possible to obtain an appreciably greater half-width for the kinetic energy distributions of the fission fragments from Cf^{252} than from U^{236}.

The dependence of the first term, R_0, on the mass ratio of the fragments must be determined from an experimental yield curve. If one specifies the value of R_0 for a specific mass ratio, then from the yield curve, values of R_0 can be determined for all other mass ratios. In this way one can obtain a family of R_0 curves consistent with the yield curve. One curve from this family was selected for the thermal neutron fission of U^{235} which gave approximately the correct average kinetic energy of the fragments at the center of the peak in the mass yield curve. This R_0 curve is the solid line in Fig. 1. The fit to the mass yield distribution given by

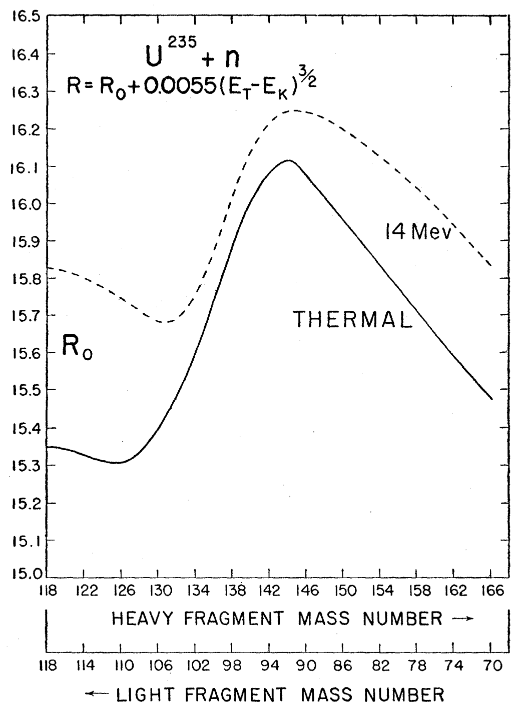

FIG. 1. Plots of the first term, R_0, in fermis, in the expression for the radius of separation of the fission fragments, as empirically determined for various mass ratios in the thermal and 14-Mev neutron fission of U^{235}.

this R_0 curve is shown in Fig. 2. The calculated yield refers to the primary yield before neutron emission from the fragments. The experimental data are chemical yield measurements[9] which are displaced to the left of the primary curve by the neutron emission.

It has been very gratifying to see the smoothness of the derived R_0 curve. If the level density formula of T. D. Newton[10] had been used here, the R_0 curve would have had fairly large fluctuations in it. This results from the fact that Newton's level density formula has rather crudely determined shell corrections. The R_0

[6] A. G. W. Cameron, Chalk River report CRL–41.
[7] J. S. Fraser and J. C. D. Milton (private communication).
[8] J. C. D. Milton (private communication).
[9] E. P. Steinberg and L. E. Glendenin, Proc. Intern. Conf. Peaceful Uses of Atomic Energy, Geneva **7**, 3 (1956).
[10] T. D. Newton, Can. J. Phys. **34**, 804 (1956).

curve peaks at the high mass ratio side of the fission yield flat-topped peak. It decreases smoothly as one goes toward symmetry past the other side of the flat-topped peak, and it has a considerably smaller value for mass ratios near unity. The fission yield curve has a flat top because the side near symmetric division corre-responds to forming fission fragments in the vicinity of the doubly-magic nucleus Sn^{132}. The extra energy release in this region causes an increase in the calculated yield.

The procedure which has been outlined has value only if it gives correct predictions about fission properties which have not been put into it. In particular, it should predict the average kinetic energies for mass ratios other than the chosen one at the center of the peak in the yield curve, and it should predict independent yields correctly.

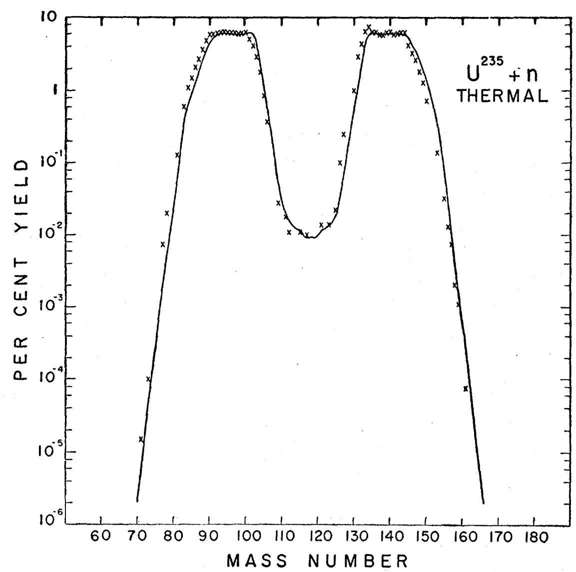

FIG. 2. The mass yield curve corresponding to the solid line in Fig. 1, compared with the best chemical yield determinations as compiled by Steinberg and Glendenin.

The predictions of the average kinetic energy for different fission modes are compared with the time-of-flight measurements of Stein[11] in Fig. 3. There are some systematic deviations between the experimental points and the curve. It is at present difficult to assess the significance of these deviations because there is lack of good agreement among various experimental measurements of these kinetic energies. In general, however, there appears to be a fair amount of agreement between the trends of the line and the points in Fig. 3. This is the feature which must be satisfied before one can have assurance that the R_0 curve gives real information about the separation of charge centers at the moment of scission. The lack of such agreement would indicate

[11] W. E. Stein, Phys. Rev. **108**, 94 (1957).

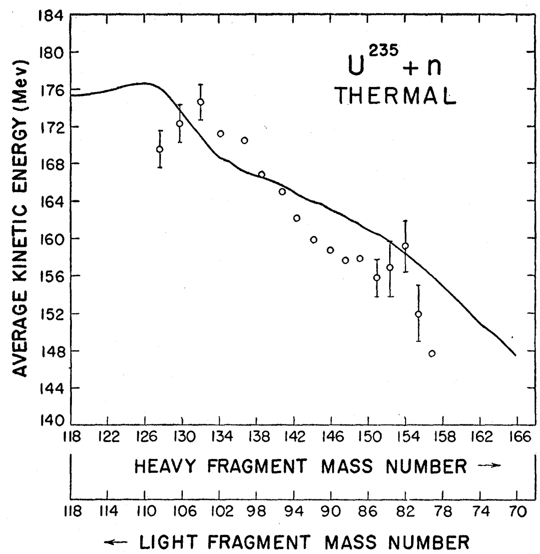

FIG. 3. The average kinetic energy as a function of fission mode compared with the time-of-flight data of Stein.

that the R_0 curve is simply a parameter which compensates for variations in internal reduced widths.

The probability distributions of kinetic energy are shown for five selected fission modes in Fig. 4. The middle three curves are in the peak of the mass yield curve, the right-hand curve is for completely symmetric fission, and the left-hand curve is in the wings of the mass yield distribution. The fission modes were selected to represent maximum calculated primary yields along the beta decay chains at the indicated mass numbers. The half-widths of these curves are in the vicinity of 14.5 Mev near the peak in the mass yield curve, in agreement with observation. The left-hand sides of these curves have very similar shapes; this results from a smooth variation in the product of a very large probability corresponding to the large number of nuclear levels available when the fragments are highly excited,

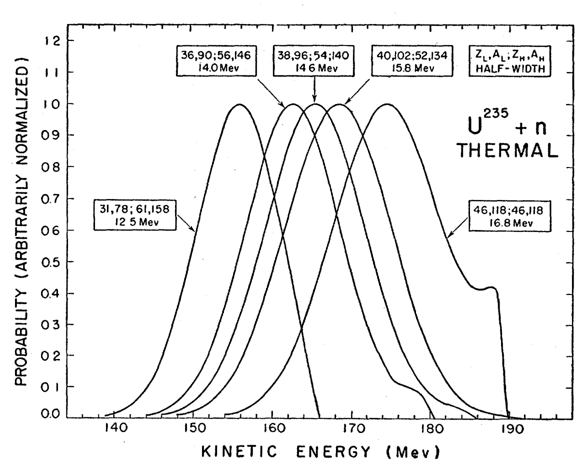

FIG. 4. The probability distributions of kinetic energies for five selected fission modes as computed by the method of this paper.

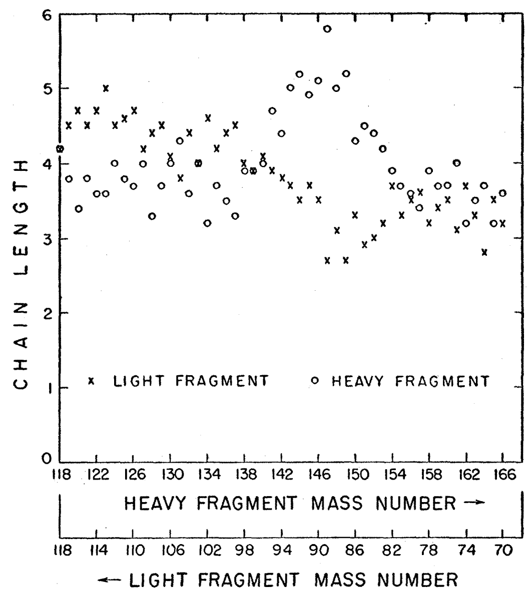

FIG. 5. The chain lengths predicted by the calculations. The equal-chain-length hypothesis requires that the circles and crosses should be intermingled.

and the very small probability characteristic of a very deep penetration through the Coulomb barrier. On the right-hand side of the curves the barrier penetration probability is high, but the fragments are formed with low excitation energies at which there is greater dependence of the level densities on shell effects. Therefore the right-hand sides of these curves vary considerably from one curve to another. There is some experimental evidence for the smoothness of these probability distributions on the low-kinetic energy sides and for more complicated features on the high-kinetic energy sides.

Direct comparisons of the independent yield predictions with experiment are difficult because calculations of neutron emission from the primary fragments have not yet been made. The comparison is easiest when applied to fission fragments corresponding to low yields on the low charge side of a given mass chain. Such fragments emit neutrons, but the yields fall sufficiently steeply with decreasing primary charge that it is not necessary to consider yield increments from neutron emission by more massive fragments. The only experimental information on such yields is the recent beautiful work of Wahl,[12] who has measured independent yields of many krypton and xenon isotopes. It appears that this calculation has underestimated the yields of the heaviest krypton isotopes and overestimated the yields of the heaviest xenon isotopes by factors of the order of 3. When one considers all the sources of error which can come into this calculation from uncertainties in energy releases and level densities, this disagreement does not appear to be serious.

Another way of checking independent yields is to examine the predictions of this calculation regarding the chain lengths which correspond to the formation of the most probable fission mode of a given mass. According to the equal-chain-length hypothesis,[13] the number of beta-decay steps between the fragment of maximum yield and the center of the valley of beta stability should be equal for light and heavy complementary mass chains. The chain lengths corresponding to the maximum predicted primary yields were computed by assuming the center of the valley of beta stability to be the line drawn on the General Electric Chart of the Nuclides.[14] These chain lengths are plotted in Fig. 5. It may be seen that the equal chain length hypothesis requires the circles and crosses to be intermingled in this figure. This requirement is fairly closely satisfied except in the region where the heavy fragment contains nearly a closed shell of 82 neutrons. This is essentially in agreement with the experimental evidence.

It is of interest to examine the reasons for the success of the equal-chain-length hypothesis. Since the valley of beta stability in the mass surface has a smaller curvature in the heavy region than in the light, the fission modes with maximum energy release on the mass chains correspond to larger chain lengths in the heavy region than in the light. However, the height of the Coulomb barrier is proportional to the product of the fragment charges, and this barrier height decreases fairly rapidly as the charge splitting becomes more asymmetric, except for mass ratios near unity. Thus the fission modes of greatest energy release on a mass chain can be slightly surpassed in yield by neighboring modes of greater charge asymmetry owing to the larger barrier penetration probabilities of the fragments in the latter modes. The greater asymmetry of charge division thus tends to equalize the chain lengths.

Some preliminary work has been done on the variation of the radius parameters necessary to fit data on nonthermal neutron fission of U^{235} and on the fission of other nuclei. This will be described here only briefly. The data on the fission of U^{235} with 14 Mev neutrons is somewhat hard to interpret because of the uncertainty in the amount of U^{236} excitation energy at the moment of fission. If it is assumed that the neutron and fission widths are about equal in this nucleus, then the R_0 curve necessary to fit the yield and average kinetic energy data is approximately given by the dashed line in Fig. 1. It is interesting to note that the entire R_0 curve has been raised relative to that for thermal fission, perhaps indicating that the additional fuzziness of the nuclear surface associated with higher excitation energy allows the nuclear system to attain a greater

[12] A. C. Wahl (to be published).

[13] Glendenin, Coryell, and Edwards, *Radiochemical Studies: The Fission Products*, Natl. Nuclear Energy Ser., Div. IV, edited by C. D. Coryell and N. Sugerman (McGraw-Hill Book Company, Inc., New York, 1951), Vol. 9, p. 489.

[14] J. R. Stein and E. F. Clancy, Knolls Atomic Power Laboratory publication (1956).

degree of distortion before separation into fragments must take place. It is also of interest to note that R_0 has increased more for symmetric fission than for asymmetric fission.

The experimental data for the spontaneous fission of Cf^{252} show that an R_0 curve is required which peaks at a smaller mass ratio than in Fig. 1 and which falls more steeply from the peak in the direction of symmetry. The data for the fast neutron fission of Ra^{226} indicate that an R_0 curve is required which peaks at a greater mass ratio than in Fig. 1 and which probably falls less steeply from the peak in the direction of symmetry. These preliminary analyses suggest that there are systematic trends in the properties of the R_0 curve as one passes from one nucleus to another, but much more work remains to be done before such trends can be established in detail.

The general conclusions concerning nuclear sizes which can be drawn from this work are that there tends to be an expansion in the nuclear surface with excitation energy which is probably not closely correlated with a volume expansion coefficient, and that there is a smaller radius of separation into fragments in symmetric fission than in asymmetric fission.

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Interpretation of Scattering Cross Sections in Terms of Nuclear Size

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1. INTRODUCTION

As experimental measurements of scattering cross sections are made with greater and greater accuracy, so our ideas of interpreting them in terms of nuclear size have to be continuously refined. Thus while only a few years ago we referred to something vaguely called the nuclear radius R, which obeyed approximately the law,

$$R = r_0 A^{\frac{1}{3}}, \tag{1}$$

we now distinguish a halfway radius c from a root-mean-square radius $[\langle r^2\rangle]^{\frac{1}{2}}$, we investigate a surface transition region and the relative density of neutrons and protons in it, we try to relate the density distribution to an optical model potential and we attempt to fit experimental proton-nucleus cross sections of such complexity that even the variation of an arbitrary function and four arbitrary constants is not enough to obtain a good fit.

Let us summarize the deductions that can be made from the experimental results.

A. The Stanford electron scattering experiments show that the proton distribution is characterized by a central region of constant density and a surface region in which the density drops to zero, which within quite narrow limits ($\pm 5\%$) is of constant thickness for all spherical nuclei. Because of the extreme insensitiveness of the electrostatic potential to small variations in the charge density, no information can be obtained about the functional form of the proton density distribution.[1]

B. Nucleon-nuclear scattering experiments in the 0–90-Mev region yield information about the parameters of the optical model potential. The complexity of the results is matched here by the complexity of the parameters, some of which are certainly energy dependent. All that can be said at present is that although there is no single set of parameters that fits the experimental results better than all others, the optical potential would appear to extend beyond the distribution of nuclear matter.[2,3]

C. The absorption cross sections for pions and nucleons by nuclei in the Bev region give the most direct information about nuclear size. They have shown that there is no significant difference between the neutron and proton distributions.[4]

2. ANALYSIS OF ASSUMPTIONS MADE

It is now necessary to point out two conclusions which have been drawn erroneously by other investiga-

[1] R. Hofstadter, Revs. Modern Phys. **28**, 214 (1956).
[2] Bjorklund, Blandford, and Fernbach, Phys. Rev. **108**, 795 (1957); A. E. Glassgold and P. J. Kellogg, Phys. Rev. **107**, 1372 (1957), and private communication; R. Jastrow and I. Harris, O.N.R. Decennial Symposium 1957.
[3] L. R. B. Elton, Nuclear Phys. **5**, 173 (1958).
[4] Abashian, Cool, and Cronin, Phys. Rev. **104**, 855 (1956).