Correlation of Fission-Fragment Kinetic-Energy Fine Structure with a Semiempirical Mass Surface*

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A correlation has been established between fine structure in primary fragment yields for thermal-neutron-induced fission of several targets and structure in the energy release calculated from semiempirical mass equations. The origin of the structure in the calculated energy release has been elucidated and can be attributed to two general properties of the mass surface: (1) The energy release for even-even products is greater than for odd-mass products, so that the structure is determined by the mass surface for even-even products. (2) The structure for even-even products has a periodicity of about 5 mass units because the mass number of the most stable nuclide for a given Z changes about 2.5 units for a unit change in Z. The existence of structure in the fission fragment yields suggests that there is a preference for forming even-even primary fragments in the fission of an even-even compound nucleus.

I. INTRODUCTION

FINE structure has been observed in the kinetic-energy spectra of fission fragments from thermal-neutron-induced fission of U^{233}, U^{235}, and Pu^{239},[1-4] and from the spontaneous fission of Cf^{252}.[5,6] It has been pointed out that there is a correlation between this experimentally observed structure and structure in a semiempirical mass surface.[3,7] It is our purpose to consider this correlation in detail and to inquire into its origin.

II. THE STRUCTURE AND THE CORRELATION

The most striking way of presenting the experimental results[1,4] is a plot of the kinetic-energy spectrum of heavy-fission fragments in coincidence with light fragments of a fixed energy. Such a plot is shown in Fig. 1.

Structure as a Function of Heavy Fragment Mass

The measured light and heavy fragment energies define the masses of the fragments according to the relationships

$$A_1/A_2 = E_2/E_1,   \qquad (1)$$

and

$$A_1 + A_2 = A   \qquad (2)$$

(if we ignore neutron emission). Here, A_1 and A_2 are the mass numbers of the two fission fragments, E_1 and E_2 are their respective energies, and A is the mass number of the fissioning nucleus. Thus each curve in Fig. 1 can be transformed to a new coordinate system of counts per unit mass as a function of the mass of either the heavy or light fragment. Results of such transformations are shown in Fig. 2 for the fissioning systems $U^{235}+n$, $U^{233}+n$, and $Pu^{239}+n$. The most striking feature of these data is that for all three systems the peaks appear at practically the same heavy fragment mass numbers, namely, 135, 141, and 146 (with less distinct peaks or shoulders, occuring at about mass numbers 151 and 156 in each case). The time-of-flight experiments of Milton and Fraser[2,3] show the same effect; that is, at high values of the kinetic energy there is a higher probability for divisions in which the heavy fragment mass is 134, 140, and 146 than there is for other divisions. For Cf^{252}, the maxima are reported to be at heavy fragment masses 140, 146, and 152.[5] The periodicity of the structure is about 5 mass units.

It would be preferable to take neutron emission into account in doing the transformations that lead to the curves shown in Fig. 2. However, for the three fissioning nuclei under consideration we know only the average number of neutrons emitted as a function of the mass number of the fission fragments. We need to know the average number of neutrons as a function not only of the fragment mass but also of the total kinetic energy.

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[1] W. M. Gibson, T. D. Thomas, and G. L. Miller, Phys. Rev. Letters 7, 65 (1961).

[2] J. C. D. Milton and J. S. Fraser, Phys. Rev. Letters 7, 67 (1961).

[3] J. C. D. Milton and J. S. Fraser, Can. J. Phys. 40, 1626 (1962).

[4] T. D. Thomas, W. M. Gibson, G. J. Safford, and G. L. Miller (to be published).

[5] J. S. Fraser, J. C. D. Milton, H. R. Bowman, and S. G. Thompson, Bull. Am. Phys. Soc., 8, 370 (1963).

[6] S. L. Whetstone, Jr., Phys. Rev. 131, 1232 (1963).

[7] R. Vandenbosch and T. D. Thomas, Bull. Am. Phys. Soc. 7, 37 (1962); and Brookhaven National Laboratory Report BNL-5876 (unpublished).

Using one or another of several assumptions about the neutron emission we can investigate the magnitude of error introduced by the use of Eq. (1) rather than a more exact expression. Terrell[8] has given a formula for the difference ΔM between the primary mass and the apparent mass resulting from the use of Eqs. (1) and (2).

$$\Delta M_1 = (M_2\nu_1 - M_1\nu_2)/A, \qquad (3)$$

where the M's refer to the masses of the fragments and the ν's to the number of neutrons emitted per fragment. Using this expression and Terrell's data on the average number of neutrons as a function of mass number,[8] we can show for fission of U^{236} that, on the average, true masses of 135, 141, and 146 will give apparent masses of 134.4, 140.8, and 146.3.

The curves shown in Fig. 2 are for events occurring with higher kinetic energies, lower excitation energies, and, hence, fewer emitted neutrons than the average. The work of Bowman, Milton, Thompson, and Swiatecki[9] on the fission of Cf^{252} indicates that the relationship between the number of emitted neutrons and the mass number of the fission fragments is qualitatively the same for all kinetic energies of the fragments, with the absolute number of neutrons decreasing by about 1 for every 6.6-MeV increase in kinetic energy. If the situation for U^{236} is similar to that Cf^{252} then the apparent masses shown in Fig. 2 should be closer to the true masses than is indicated in the above paragraph. The most unfavorable situation which is likely to occur is one in which there is enough excitation energy to evaporate one neutron, either from one fragment or the other. For
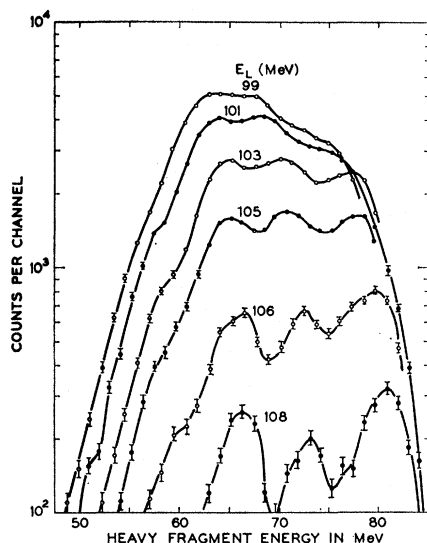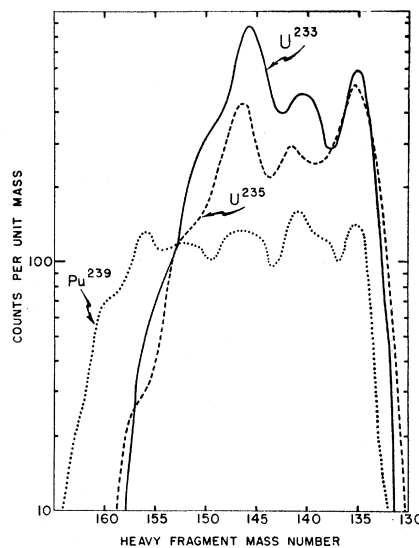

FIG. 2. Experimentally observed fine structure for the thermal-neutron induced fission of U^{233} (solid curve), U^{235} (dashed curve), and Pu^{239} (dotted curve). From Refs. 1 and 4. The data have been transformed to a coordinate system of counts per unit mass as a function of the mass of the heavy fragment. The light fragment kinetic energies are 108.7 MeV for U^{235}, 109.7 MeV for U^{233}, and 113.3 MeV for Pu^{239}.

mass numbers of interest, the largest shift that could be produced by one neutron would be if that neutron were emitted from a very light fragment, mass 80, for instance. For U^{236}, the shift in the primary heavy fragment mass, 156, would be $+0.66$ mass units.

We can conclude, therefore, that the use of Eqs. (1) and (2) to calculate the mass of the fission fragments will give an apparent mass that differs on the average by less than one mass unit from the true prompt mass. It is possible, however, as Terrell[8] has pointed out, that minor variations in the number of emitted neutrons from one mass number to the next "can create apparent peaks of mass yield where none exist, or can eliminate true peaks in mass yield." We cannot exclude the possibility that the data of Figs. 1–2, 4–6 have been so distorted. However, there is good agreement between the positions of the maxima as determined by the solid-state counter experiments[1,4] and the positions as determined by the time-of-flight experiments.[2,3] Therefore, we are probably safe in using the solid-state counter data rather than the time-of-flight data, which have been presented in a manner that does not make the structure particularly obvious.

Excitation Energy of the Fission Fragments

Using Cameron's mass formula,[10] Milton[11] has calculated the energy released in fission for any mass and charge division of the systems under consideration. If,

FIG. 1. Experimentally observed fine structure for the thermal-neutron-induced fission of U^{235} (from Ref. 1). Curves show the energy spectra of heavy fragments in coincidence with light fragments of the indicated energy.

[8] J. Terrell, Phys. Rev. 127, 880 (1962).
[9] H. R. Bowman, J. C. D. Milton, S. G. Thompson, and W. J. Swiatecki, Phys. Rev. 129, 2133 (1963).

[10] A. G. W. Cameron, Can. J. Phys. 35, 1021 (1957).
[11] J. C. Douglas Milton, University of California Radiation Laboratory Report, UCRL-9883 Rev. (unpublished).
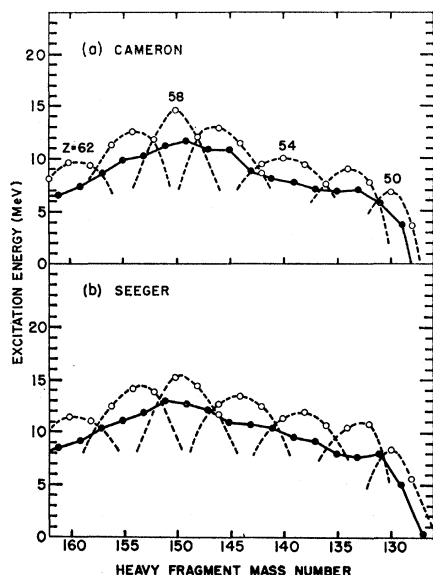
FIG. 3. (a) Total excitation energy of the fission fragments from the thermal-neutron induced fission of U^{235} for various mass and charge divisions and a light-fragment kinetic energy of 108 MeV. Open circles are for even-mass number products and closed circles are for odd-mass number products. Dashed lines connect points corresponding to a given division of charge. Solid lines connect points of maximum excitation energy for odd-mass products. Calculated from Cameron's masses. (b) Same as (a) except masses taken from Seeger.

as in Fig. 2, we specify the light fragment mass number, we can, using Eqs. (1) and (2), calculate the heavy fragment kinetic energy, the total kinetic energy, and the light fragment mass number. The final excitation energy of the fragments is the difference between the total energy released, as calculated by Milton, and this total kinetic energy. For each mass division, there will be a range of possible excitation energies, corresponding to the different possible charge divisions.

The results of such a calculation for U^{235} plus thermal neutrons are shown in Fig. 3. The maximum excitation energy for even-even fragments are shown as open circles and for odd-mass fragments as closed circles. The upper curves show the results based on Cameron's formula, as calculated by Milton,[11] while the lower curves show results based on Seeger's formula.[12]

The procedure outlined above gives the excitation energy for *prompt* fragments having a given light fragment kinetic energy and heavy fragment mass number. To make a comparison of the experimental data with the calculated excitation energy curves we must adjust one or the other for the effects of neutron emission. If the average number of neutrons is emitted from these fragments, then the excitation energy curve shown in Fig. 3 should be lowered by about 1 MeV at the high-mass end and about 2 MeV at the low-mass end. Since the number of emitted neutrons is less than the average, the correction is somewhat smaller. The neglect of this

[12] Philip A. Seeger, Nucl. Phys. 25, 1 (1961).

correction does not introduce an appreciable error in the position along the abscissa of the various curves shown in Fig. 3.

We will consider the source of the fluctuations in more detail in Sec. III. We note here that they are not the result of any unexpected irregularities in the mass surface, but arise primarily because of two factors: (1) The energy release for even-even products is greater than for odd-mass products, so that the structure is determined by the mass surface for even-even products. (2) The structure for even-even products has a periodicity of about 5 mass units because the mass number of the most stable nuclide for a given Z changes about 2.5 units for a unit change in Z. Shell effects also play a role, as can be seen in the high-excitation energy for $Z=58$ and $A=152$ (50 neutrons in the light fragment) and in the falloff of excitation energy for heavy fragments with mass less than 132 (50 protons and 82 neutrons). These factors combine to produce the variations seen in Fig. 3. However, it must be pointed out that, if it were not for odd-even effects due to pairing, the energies released for fission to even-even products would be practically no different from those released for fission to odd-mass products. As can be seen from the solid circles in Fig. 3, the excitation energy curve for odd-mass products is rather featureless.

Thus, although the structure in the mass surface does not arise from the pairing effect, if there were no pairing effect, the structure in the even-even mass surface would be completely obscured by the odd-mass and odd-odd mass surfaces. Furthermore, the application of a statistical theory with the usual assumption of measuring level densities from a reference surface corresponding to odd-mass or odd-odd nuclei will fail to reproduce the observed structure.
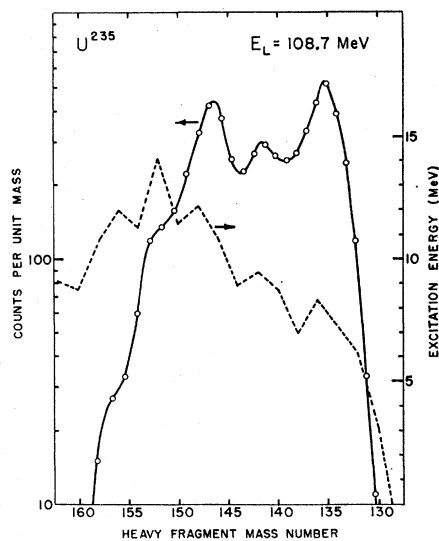

FIG. 4. Correlation between structure observed for U^{235} and the calculated maximum excitation energy. Data from Refs. 1 and 4.

Another feature to be noted is that although the total energy release for this system peaks at heavy fragment mass 132, the excitation energy peaks at mass 152. In this particular representation of the data, the total kinetic-energy release increases rapidly as the mass number of the heavy fragment decreases. It is this increase that brings about the shift of the position of the highest excitation energy from mass 132 to 152.

Correlation of Mass-Surface Structure With Kinetic-Energy Structure

A comparison of the calculated maximum excitation energies with the experimental data is shown in Figs. 4, 5, and 6 for the three systems. We see that there is a correlation between the structure in the data and the structure in the energies. It is clear that as we go to higher kinetic energies, it will be energetically impossible to produce fragments with certain mass numbers even while it is still possible to produce those with only slightly higher and slightly lower mass numbers. At lower kinetic energies, or higher excitation energies, the structure will tend to disappear because the small energy differences will have less influence on the fragment yield. It is interesting to note, however, that some structure does persist even in the most probable modes of fission, as shown in Fig. 1. The structure may be even more pronounced at the more probable modes than is indicated in Fig. 1, since dispersions due to neutron emission will tend to wash out the structure at lower total kinetic energies.

We see also from these curves that for the examples considered the fragments have fairly low excitation energies—not more than about 7 MeV per fragment. In general, therefore, there will be no neutrons emitted from these fragments.
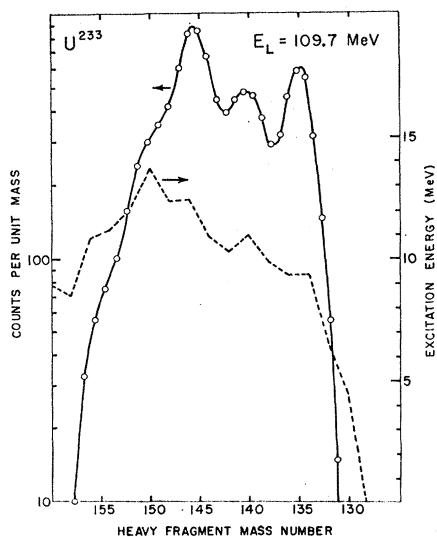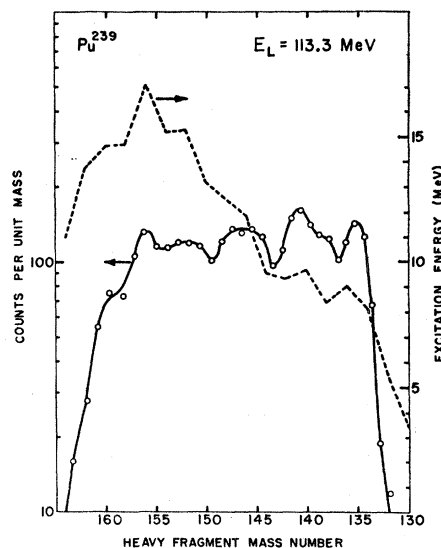

Fig. 6. Correlation between structure observed for Pu[239] and the calculated maximum excitation energy. Data from Ref. 4.

Effects of Mass-Dependent Pulse-Height Defect

The energy calibration upon which the curves shown in Figs. 1, 2, 4, 5, and 6 are based was calculated under the assumption that there is one linear pulse height versus energy curve for all masses of fission fragments. The details of the calibration method will be described in Ref. 4. Because there is in fact a mass-dependent pulse-height defect, we should use a family of calibration curves, one for each mass, rather than a single curve.[13] The data that are now available on this question are not sufficient for us to construct such a family of curves.

We have tried to estimate how inclusion of a properly mass-dependent pulse-height defect would affect the curves shown in Fig. 4 as an example. We have come to the following conclusions: (1) The experimental curve should be shifted to slightly lower mass numbers; for instance, the peak at 146.5 mass units should be at 146.0, and the peak at 135.5 should be at 135.3. (2) The light fragment kinetic energy is not constant at 108.7 MeV, but ranges from about 107.5 MeV at heavy fragment mass 160 to about 112 MeV at mass 130. (3) The excitation energy curve should be about 2 MeV higher at heavy fragment mass 160 and about 6 MeV lower at mass 130.

The net effect is to leave the correlation unchanged. In one or two cases this correction would cause a peak in the experimental data to be shifted further from the associated peak in the excitation energy curve. In one or two other cases the two peaks are shifted closer together. For the most part, however, there is no perceptible change.

Fig. 5. Correlation between structure observed for U[233] and the calculated maximum excitation energy. Data from Ref. 4.

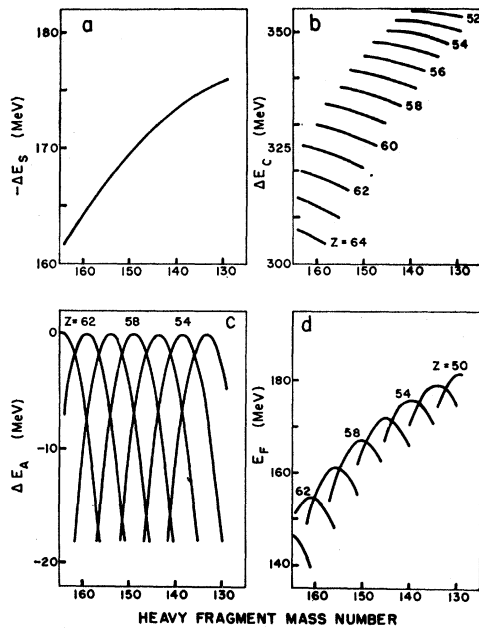[13] H. C. Britt and H. E. Wegner, Rev. Sci. Instr. 34, 274 (1963).

FIG. 7. Surface (a), Coulomb (b), asymmetry (c), and total (d) energy changes for the fission of U^{236}, as calculated from a semi-empirical mass formula.

III. SOURCE OF MASS SURFACE STRUCTURE

As we have indicated above, the fluctuations in the excitation energy or energy release do not arise from any unexpected irregularities in the mass surface. It is interesting to inquire further into just what their source is.

We take the semiempirical mass equation given by Evans[14] to define the mass surface. Although this equation is somewhat simpler than those used by Seeger[12] and by Cameron,[10] the basic structure of the three equations is the same. The general behavior of the various terms can be illustrated most easily with Evans' equation and the conclusions drawn can be applied to the other equations with only slight modification. The energy E_f released in a particular mode of fission is given as

$$E_f = A_c \left[\frac{Z^2}{A^{1/3}} - \frac{Z_1^2}{A_1^{1/3}} - \frac{Z_2^2}{A_2^{1/3}} \right] + A_s (A^{2/3} - A_1^{2/3} - A_2^{2/3})$$

$$+ A_a \left[\frac{(A-2Z)^2}{A} - \frac{(A_1 - 2Z_1)^2}{A_1} \right.$$

$$\left. - \frac{(A_2 - 2Z_2)^2}{A_2} \right] + \Delta. \quad (3)$$

The three coefficients A_s, A_c, and A_a are the surface, Coulomb, and asymmetry coefficients respectively, and Δ is a term to represent shell and pairing effects. The

[14] Robley D. Evans, The Atomic Nucleus (McGraw-Hill Book Company, Inc., New York, 1955), pp. 365–383.

behavior of each of the first three terms is shown in Figs. 7(a), (b), and (c), respectively, and their sum in 7(d) for the fissioning nucleus U^{236}.

We see that the major part of the energy comes from the difference between the Coulomb and surface terms and that the oscillations arise from the asymmetry term. The term Δ has two effects. First, it causes divisions leading to fragments with closed shells to be more energetically favorable than would be otherwise expected. Second, it causes the energy release surface for odd-mass fragments to be depressed somewhat below that for even-even fragments.

The curves of Fig. 7(c) can be shown to have their maxima at a mass number such that

$$Z_1/A_1 = Z_2/A_2 = Z/A.$$

The slopes of the curves for the Coulomb and surface energies are such that these maxima are shifted by about 1 mass unit to higher mass numbers in Fig. 7(d). If we subtract the kinetic energy from the curves in Fig. 7(d), there is a further shift of about 2 mass units, with the net result that the excitation energy maxima occur at mass numbers about 3.5 mass units higher than do the asymmetry energy maxima.

Since the structure in the excitation energy curves is determined by the even-even products, we may expect to find maxima every $2A/Z$ mass units (≈ 5), inasmuch as the successive maxima of the asymmetry energy curves are A/Z mass units apart. The positions of these maxima should, therefore, be given by the expression

$$A_1^{\max} \approx Z_1(A/Z) + 3.5.$$

If we calculate values of $A_1^{\max}$ using this formula and compare them with the positions of the maxima in the experimental curves, we find fairly good agreement. The root-mean-square difference between the experimental and calculated values is 0.9 mass unit.

We should note that the exact position of the peaks in the excitation energy curves will depend slightly on what parameters are used for the mass equation. The period of the oscillations, 5 mass units, is, however, almost completely independent of the parameters chosen.

IV. EFFECTS OF NUCLEAR DEFORMATION

Ordinary semiempirical mass equations, such as those used above, give the masses for nuclei (usually spherical) in their ground state. However, the fission fragments, when formed, are highly distorted, and we should consider additional terms to calculate the mass of the deformed nucleus in its lowest possible energy state. We need to know what effect, if any, the deformation will have on the excitation energy fluctuations calculated above.

Any such corrected mass equation must still contain an asymmetry term, leading to a basic structure with a period of 5 mass units in the excitation energy curves

for the deformed nuclei. It is possible that the other terms in such an equation might vary rapidly enough with changes in Z and A either to shift the positions of the maxima or even to change their periods.

To approximate the effects of deformation, we have assumed that the newly formed fission fragments are prolate spheroids with collinear major axes. The expression for calculating the Coulomb and surface energies of a prolate spheriod is given by Swiatecki.[15] We assume that there is no change in the asymmetry energy with deformation. The difference between the mass of the deformed and undeformed fragments is then the deformation energy.

There is also an energy of interaction between the two fragments, that is, the energy due to the Coulombic repulsion between them. This energy is assumed equal to the total kinetic energy of the separated fragments. For any given pair of fragments having specified Z_1, Z_2, A_1, A_2, and total kinetic energy, we have chosen the major to minor axes so as to give the required interaction energy and the minimum deformation energy. The interaction energies have been calculated using the formulas given by Cohen and Swiatecki.[16] In calculating these energies, we have assumed that the distance between the centers of mass of the two spheroids is 10% greater than the sum of their major semiaxes. Such an adjustment of the distance is necessary to give reasonable values of the kinetic energy.[3,17]

Either one of two further assumptions may also be made. The first is that all shell structure has been destroyed in the deformed fragments. Then the total mass is given by the simple liquid drop mass without shell corrections plus the deformation energy. The alternative assumption is that the shell structure is not destroyed by the deformation. In this case the total mass is given by an equation such as Cameron's which includes shell corrections,[10] plus the deformation energy. It is necessary, however, in the latter case, to take into account the differing deformability of the various nuclides, depending on whether they are close to or far from closed shells. To include this effect, we have used the method described by Vandenbosch[17] and deformability parameters that he has determined.

The results of calculations on the basis of these assumptions are shown in Fig. 8, where the upper part of the figure is based on the assumption that the shell structure has been destroyed by the deformation and the lower part on the contrary assumption. In each case we show the excitation energies for the undeformed nuclei and those for the deformed.

Two features of these curves are to be noted. First, in both cases there is practically no change in the positions
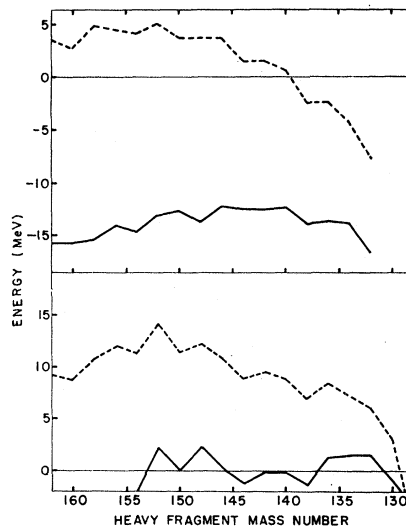

FIG. 8. Excitation energy of deformed (solid curves) and undeformed (dashed curves) fission fragments as a function of the heavy fragment mass. The upper set of curves is calculated under the assumption that the shell structure is destroyed by the deformation and the lower under the assumption that it is not.

of the maxima in the curves due to inclusion of the deformation energy. Second, the excitation energy for the very asymmetric fragments is reduced much more by the effect of deformation than is that for the more symmetric ones. This effect provides a possible explanation for the fact that the extremely asymmetric fragments, although favored by the total available energy, do not contribute to the spectra shown in Figs. 1 and 2. The deformation energy of these fragments is so high at the scission point that it exceeds the total energy available for excitation. Thus, these modes of fission can take place only by barrier penetration.

V. CONCLUSION

A correlation between structure in fission yields and structure in a semiempirical mass surface has been established. The structure in the semiempirical mass surface is not due to any special irregularities in the mass surface, but is a consequence of two rather general properties of the mass surface: (1) The energy release for even-even products is greater than for odd-mass products, so that the structure is determined by the mass surface for even-even products. (2) The structure for even-even products has a periodicity of about 5 mass units because the mass number of the most stable nuclide for a given Z changes about 2.5 units for a unit change in Z. While the period of the structure is a general property due to the form of the mass equation, the exact positions of the peaks in the excitation energy curves depend somewhat on the details of the mass equation. The latter fact may enable more critical tests of proposed mass equations.

[15] Wladyslaw J. Swiatecki, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy* (United Nations, Geneva, 1958), Vol. 15, p. 249.

[16] S. Cohen and W. J. Swiatecki, Ann. Phys. (N. Y.) **19**, 67 (1962).

[17] R. Vandenbosch, Nucl. Phys. **46**, 129 (1963).

Perhaps the most interesting observation which can be made is that the structure persists to the more probable energies where the final excitation energy is larger. This can partly be attributed to the fact that at scission, most of what appears as final excitation energy is tied up in deformation energy. The existence of structure at the higher excitation energies implies that the primary fission fragments may be predominantly even-even nuclei. This enhancement of even-even primary fragment yields will be modified by neutron emis-

sion. Calculations based on Terrell's parameters[18] for neutron emission probability distributions indeed indicate that there are approximately equal probabilities for a primary fragment to emit an even or an odd number of neutrons. This means that the yield of even-even secondary fragments will not show this enhancement and that the structure will be obscured in the radiochemical yield measurements.

[18] James Terrell, Phys. Rev. **108**, 783 (1957).

Comparison to Experiment of the Amati-Leader-Vitale Multipion-Exchange Contributions*

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The phase shifts obtained by Amati, Leader, and Vitale (ALV), using the Cini-Fubini approach to the Mandelstam representation, are compared to proton-proton scattering data at 51.8, 96.5, 142, 210, and 310 MeV. The ALV phases yield a considerably better fit to the data than do pure one-pion exchange, but considerably poorer than modified phase analyses. The unusual behavior of the ALV $^3F_2$ and ϵ_4 is not confirmed. The lower angular momentum phases from ALV+data are generally not greatly different from those of modified phase analyses.

I. INTRODUCTION

IN a recent series of papers, Amati, Leader, and Vitale[1] (hereafter referred to as ALV) have applied the Cini-Fubini approach (to the Mandelstam representation) to the problem of nucleon-nucleon scattering. Within this framework, ALV were able to obtain predictions for the *total* (correlated and uncorrelated) two-pion exchange contribution for nucleon-nucleon phase shifts with orbital angular momentum $L \geq 2$ and incident laboratory energy $E_L \leq 300$ MeV. ALV added in one-pion exchange (OPE) and three-pion exchange (ω) as pole terms, and computed the resulting phase shifts. Those phases were then compared to YLAM and SMMN curves from energy-dependent phase-shift analyses of experimental data. The predicted "theory" curves in the most recent ALV reference show that the ALV multipion contributions generally correct OPE toward the "experimental" YLAM and SMMN curves.

The present work was motivated by two observations: (1) There are now more experimental data available than when the YLAM fit was made. This may also be true of the SMMN fit. (2) The SMMN curves have no errors shown, so there is no way of estimating how closely they should be matched by theoretical predictions. This is remedied here by computing the standard

deviations for the phase shifts deduced from the experimental data, and by fitting the ALV phases directly to the data.

Section II specifies the data used, and Sec. III the method of analysis. IV defines the modified phase-shift analyses used for comparison. In Sec. VA the ALV fit to the data is compared to that of the pure one-pion exchange, and to the fits of the modified phase analyses. The extent to which the result depends on the $J=2$ phases is examined in Sec. VB. Finally, in Sec. VC, other current two-nucleon models are examined for the strange energy dependence of the ALV $^3F_2$ and ϵ_4. Also, an attempt is made there to confirm or refute the behavior by data analysis.

II. DATA USED

A compilation of the data which were used is shown in Table I. There were 222 pieces of proton-proton scattering data near 51.8, 96.5, 142, 210, and 310 MeV. All of the data were treated as though measured at the nearest energy in the above list. This probably did not introduce a significant amount of error, since the (absolute) cross section and polarization normalizations were treated as data separate from the relative angular distributions, and the shapes of angular distributions do not change rapidly with energy. Small-angle cross-section shapes probably are more rapidly varying with energy, but the only forward-angle cross sections included in this work were used at the measured energies.

[1] D. Amati, E. Leader, and B. Vitale, Phys. Rev. **130**, 750 (1963), and previous publications cited therein.