Interpretation of the Fragment Mass Distribution for U^{235} Thermal-Neutron-Induced Fission with Long-Range Alpha Emission

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An analysis of the fragment mass distribution for the thermal-neutron-induced ternary fission of U^{235} is given. Quantitative comparison of the mass-distribution data for binary and ternary fission is consistent with the influence of the closed-shell configurations in fission, although it is shown that these mass distributions of themselves are not sufficient to determine unambiguously the origin of the alpha particle. Results of this analysis are discussed in relation to some recently proposed models of ternary fission. It is shown that the results of measurements of the number of neutrons emitted in binary fission, as a function of fragment mass, are consistent with the cluster-model interpretation.

INTRODUCTION

An analysis of the fragment mass distributions[1] in thermal-neutron-induced binary and ternary (where the third particle is a long-range alpha particle) fission of U^{235} has been carried out. It is the purpose of this short paper to discuss quantitatively the conclusions which may be drawn; these conclusions are then discussed in relation to some recently-proposed models (or mechanisms) for binary and ternary fission. It appears, in particular, that the cluster model[2] contains the essential features necessary for a consistent picture of the process and is in many respects a quite general model, as we shall see. Results of recent measurements[3] of the number of neutrons emitted per fission, as a function of fragment mass near symmetric fission, are shown to be consistent with the cluster-model interpretation.

ANALYSIS

In Fig. 1(a) are shown the mass distribution for binary fission and the mass distribution for ternary fission normalized for comparison of shapes. Consider these mass distributions in detail. If we first assume, for purposes of discussion, that fission with alpha emission is a rapid two-step process, with alpha-particle emission from the fragment of initial mass M, then the yield $Y_T(M-4)$ in ternary fission of mass $M-4$ is given by

$$Y_T(M-4)=Y_B(M)P_\alpha(M)\,, \qquad (1)$$

where $Y_B(M)$ is the binary-fission yield of mass M, and $P_\alpha(M)$ is the probability of alpha-particle emission from initial mass M. The yields $Y_T(M-4)$ and $Y_B(M)$ are related to the yields of their complementary masses respectively as follows:

$$\begin{aligned} Y_T(M-4)&=Y_T[232-(M-4)]=Y_T(236-M)\,,\\ Y_B(M)&=Y_B(236-M)\,. \end{aligned} \qquad (2)$$

Therefore, $P_\alpha(M)$ may be calculated in a straight-

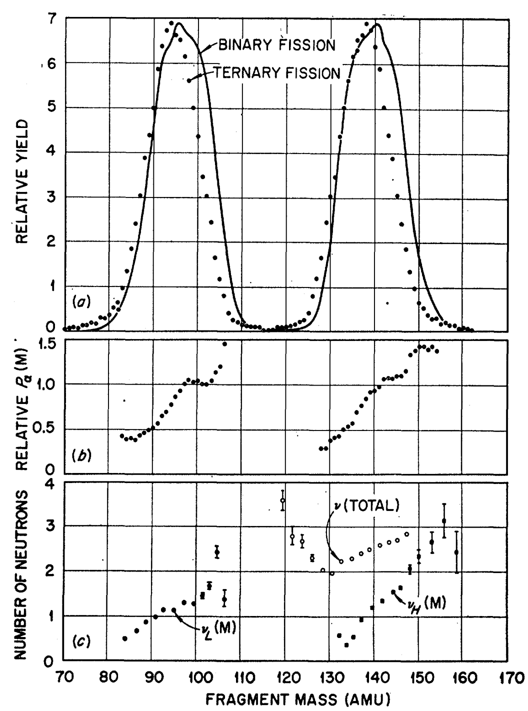

FIG. 1. (a) Fission-fragment mass distributions from energy correlation experiments (Ref. 1) for binary and ternary fission of U^{235}. (b) Relative values of $P_\alpha(M)$; see text. (c) Number of neutrons, $\nu(M)$, emitted per fission by individual fragments as a function of fragment mass (Ref. 11), and total number of neutrons emitted per fission as a function of heavy-fragment mass (Ref. 3) in the binary fission of U^{235}.

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[2] K. Wildermuth and H. Faissner, *Proceedings of the International Conference on Nuclear Structure, 1960*, edited by D. A. Bromley and E. W. Vogt (University of Toronto Press, Toronto, 1960), p. 972; Phys. Letters **2**, 212 (1962).
[3] V. F. Apalin, Y. N. Gritsyuk, I. Y. Kutikov, V. I. Lebedev, and L. A. Mikaelyan, Nucl. Phys. **38**, 193 (1962).

forward manner from the relation:

$$P_\alpha(M)=\frac{Y_T(236-M)}{Y_B(236-M)}. \tag{3}$$

Relative values of $P_\alpha(M)$ have been computed from the data of Ref. 1; these are plotted in Fig. 1(b). There are possible differences in resolution between the binary and ternary distributions, principally because of alpha-recoil effects and because of the probable differences in neutron emission. Therefore, only the relative $P_\alpha(M)$ values for masses in the ranges $83\lesssim M\lesssim 107$ and $128\lesssim M\lesssim 154$ are meaningful.

It is obvious, but important, to note that a redundancy exists in this calculation; i.e., the entire $P_\alpha(M)$ curve is not required in order to generate the experimental ternary mass-yield curve from the experimental binary mass-yield curve. For example, the ternary mass yields may be generated from the binary yields under the assumptions that $P_\alpha(M)=0$ in the range $M<118$, and that relative values of $P_\alpha(M)$ are given by the function as plotted for $M>118$. Similarly, it is possible to assume relative values of $P_\alpha(M)$ as given for $M<118$, with $P_\alpha(M)=0$ for $M>118$. An infinite number of linear combinations, point by point, of the two parts of the $P_\alpha(M)$ curve are available (on a purely formal basis from these data alone) to "explain" the relative binary and ternary mass yields.

These calculations and considerations have been made on the assumption that a rapid two-step process takes place in fission with long-range alpha emission. It is clear that the same considerations (and ambiguities) are applicable under the assumption that a single-step process (three-body breakup) occurs, provided only that the alpha particle is formed predominantly at the expense of nucleons in one or the other, but not both, of the heavier fragments.

Under the assumption that a three-body breakup occurs, an expanded development analogous to the above could be carried out to include alpha-particle formation at the expense of m nucleons in one fragment and n nucleons in the other, where $m,n=0,1,2,3,4$, and $m+n=4$. Any number of combinations of the results of such calculations would then be available to discuss the relationship between the two mass distributions; such general calculations, however, have no physical basis at present.

DISCUSSION

In the cluster model of fission, developed principally qualitatively to date, the basic process consists of the formation of two clusters in the asymmetric fissioning nucleus prior to scission.[2] These clusters consist of nucleons which compose the closed shells $Z=50$ and/or $N=82$ in the heavy fragment and $Z=40$ or $N=50$ in the light fragment, together with the excess nucleons which may separate to become part of either fragment. For cases of low-excitation energy, as we have in thermal-neutron-induced fission of U^{235}, it is energetically favorable to form these clusters in the fissioning nucleus[2]; it may be assumed, therefore, that it is energetically unfavorable to break up the closed-shell clusters once they are formed.

It was suggested in Ref. 1 that the comparison of the binary and ternary mass distributions is strong indication of the influence of closed-shell clusters in the fission process. Detailed calculations of the function $P_\alpha(M)$, shown in Fig. 1(b) would now appear to support this view quantitatively; viz., the relative values of $P_\alpha(M)$ in the regions of the closed shells, as shown in Fig. 1(b), are much smaller than those away from the closed shells. Thus, those fragments whose masses are in the region of closed shells, in particular $N=82$ or $Z=50$, essentially remain intact, independent of the binary or ternary nature of the division which occurs. This conclusion requires only the assumption that the alpha particle is formed at the expense of nucleons principally in one or the other, but not both, of the heavier fragments. No assumption is made, nor do the results determine whether a one-step (three-body breakup) or two-step process occurs; although observed angular distributions,[4–6] showing a peak at 82° with respect to the light fragment, indicate generally that at most a small fraction of the kinetic energy of the fragments is acquired at the time of alpha emission.

The one-step versus two-step nature of fission with long-range alpha emission has been the subject of recent study. In particular, Feather[7] has developed arguments in favor of the two-step hypothesis, and has extended these arguments to the proposal that all of the long-range alpha particles originate in the heavy fragment, with $P_\alpha(M)$ as given in Fig. 1(b) for $M>118$, and $P_\alpha(M)=0$ for $M<118$. Halpern[8] and Vandenbosch[9] have developed arguments which indicate that alpha emission in fission is related to the fragment deformations at scission. In either case, the influence of the stability of the closed-shall core in the fragment is apparent, the relative values of $P_\alpha(M)$ being lower in the region of closed-shell nuclei. For the case of association of alpha emission with deformation, one need only note that nuclear deformations increase from small values for closed-shell nuclei to relatively large values for nuclei removed from closed shells.

The stability of closed-shell clusters in the fragments might also be expected to influence neutron emission in fission, particularly since almost all of the emitted prompt neutrons appear to originate in the fragments

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[7] N. Feather (to be published).
[8] I. Halpern (private communication, 1963).
[9] R. Vandenbosch (to be published).

after scission.[10] In the present framework, the sawtooth character observed in binary fission[11] for the function $\nu(M)$, the average number of neutrons emitted per fragment as a function of fragment mass, is understandable: the number of emitted neutrons is small in the mass region 85-90 amu (where $N\cong 50$) and in the mass region 130–135 amu (where $N\cong 82$ and $Z\cong 50$) and increases as a function of mass away from these regions. Difficulties in obtaining data for symmetric and near-symmetric fission of U^{235} have thus far precluded a determination of $\nu(M)$ in the crucial mass region below $\sim$130 amu. However, a measurement has been carried out by Apalin *et al.*[3] whereby the total number of neutrons emitted (for both fragments) as a function of mass ratio was determined in the region of symmetric fission for U^{235}. These results are reproduced in Fig. 1(c), as are the earlier results of Apalin *et al.*[11] showing $\nu(M)$ for U^{235}. Of most significance for the present discussion are the strong increase in total number of neutrons toward symmetric fission and the definite minimum in this function for $M\cong 130$ amu. The symmetric fragments from U^{235} neutron-induced fission are removed from the closed-shell regions, have large deformations, and, since they are highly neutron-rich, should emit a relatively large number of neutrons, as indeed is observed.

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Coulomb Excitation of Cu^{63} and Cu^{65}

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Properties of levels Coulomb excited in Cu^{63} and Cu^{65} with 4- to 8-MeV alpha particles have been investigated. The $B(E2)$'s for excitation determined from the yields of gamma rays observed in singles and coincidence spectra for the (668±4)-, (961±6)-, and (1323±16)-keV levels in Cu^{63} are, respectively, (1.16±0.12), (3.49±0.34), and $(4.0\pm0.5)\times10^{-50}\, e^2\,\mathrm{cm}^4$; for the (771±7)-, (1115±11)-, and (1474±19)-keV levels in Cu^{65}, they are (1.02±0.11), (3.45±0.38), and $(4.3\pm0.8)\times10^{-50}\, e^2\,\mathrm{cm}^4$. These results give $B(E2)$'s for decay which are 13 to 16 times the single-particle $B(E2)$. This similarity of $B(E2)$'s was predicted by de Shalit for levels which result from the coupling of the odd particle to the first excited state of the even-even core. From angular correlation studies the 961- and 1115-keV levels have been assigned spin $\frac{5}{2}$ and the 1323-keV level spin $\frac{7}{2}$. Mixing ratios obtained from these measurements along with the $B(E2)$'s establish values of 0.09 and 0.4 $(e\hbar/2\,Mc)^2$ for the $B(M1)$'s of the 961- and 1115-keV transitions.

I. INTRODUCTION

Because Cu^{63} and Cu^{65} have a structure of one odd proton beyond a closed shell, a study of their level properties should provide an excellent means for testing the validity of the single-particle core coupling model. According to this model some low-lying levels can result from the coupling of the odd particle to the even-even core. With this type of model Lawson and Uretsky[1] derived a relation (the center-of-gravity theorem) between energies of states arising from the coupling of the odd particle to the core in its first excited state and of the energy of the first excited state in the neighboring even-even nucleus. For nuclei like Cu^{63} and Cu^{65} which have a ground-state spin of $\frac{3}{2}$, four excited levels with spins of $\frac{1}{2}$, $\frac{3}{2}$, $\frac{5}{2}$, and $\frac{7}{2}$ result from this coupling. Lawson and Uretsky found the center-of-gravity theorem was satisfactorily fulfilled by the first four excited levels in both copper nuclides for the proper choice of spins.

In a generalization of Lawson and Uretsky's weak-coupling model, de Shalit[2] predicted that the reduced $E2$ transition probabilities for decay, $B(E2)_d$, from the four members of the multiplet to the ground state will be equal to the $B(E2)_d$ between the 2+ and 0+ states in the adjacent even-even nucleus. de Shalit also pointed out that for pure states the $M1$ transition probabilities are forbidden. However, a small admixture of states can give appreciable $M1$ rates.[2,3]

Another approach to the single-particle core coupling model has been taken by Bouten and Van Leuven.[4] They treated levels of the odd-mass nuclei $Cu^{59,61,63,65}$ as arising from intermediate coupling between the single particle in either the $p_{1/2}$, $p_{3/2}$, or $f_{5/2}$ orbital and the core in the ground state or first or second phonon state. In a third approach Bayman and Silverberg[5] have performed an intermediate coupling calculation

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