

Bi FISSION AT VERY HIGH EXCITATION ENERGIES

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Submitted to JETP editor June 28, 1958

J. Exptl. Theoret. Phys. (U.S.S.R.) **36**, 652-657 (March, 1959)

Nuclear emulsions were used to study relative yields in Bi fission induced by 660-Mev protons as a function of the range ratio of light and heavy fission fragments, l_L/l_H , for groups of nuclei with different mean excitation energies. The dependence of fission yields on the range ratio was found to vary nonmonotonically for some nuclei. The dependence of the mean combined fragment range on l_L/l_H has been obtained. The experimental findings are accounted for on the basis of the shell structure of the product nuclei.

ALTHOUGH the mass and energy distributions of nuclear fission products have been studied extensively both experimentally and theoretically, much work remains to be done. The mass distribution differs between elements and varies with the bombarding energy. For atomic number 90 and higher at low bombarding energies fission is characteristically asymmetric, but the asymmetry diminishes with increasing mass of the fissioning nucleus,¹ and increasing excitation energy.² Very high-energy particles induce predominantly symmetric fission.³ For atomic number 83 and lower symmetric fission is predominant at both high⁴ and low⁵ excitation energies. Evidence has recently been obtained⁶ that fission of elements with Z between 83 and 90 may be even more complex in character. It is difficult to obtain a consistent picture from the large number of separate experimental facts. Various authors have attempted in different ways⁷ to account for individual fission modes, but there is no single explanation for all of the observed effects. Further investigations in this field will therefore be extremely important.

1. EXPERIMENTAL PROCEDURE AND RESULTS

The present work is a detailed study of Bi fission induced by high-energy particles. P-9 emulsions impregnated with a Bi salt were irradiated in the synchrocyclotron of the Joint Institute for Nuclear Studies by 660-Mev protons. After processing the emulsions were examined microscopically to detect Bi fission events; the horizontal and vertical projections of fragment tracks were then carefully measured and the true range of each fragment was calculated with allowance for shrinkage. The ratio of true fragment ranges was taken

as a measure of fission asymmetry. Special fine-grained emulsions permitted visual determination of the separation point of fission fragments even when unaccompanied by charged particle emission.⁸ The number of prongs associated with each fragment separation point was determined and the prongs were identified as belonging to either to singly- or doubly-charged particles. The sensitivity of the emulsions used is about 100 Mev for protons, thus permitting registration of the entire evaporation spectrum of charged particles and a considerable fraction of the knock-on protons. Most unobservable knock-on protons are associated with low excitation energy. On the other hand, at high excitation energies the nuclear cascade branches so strongly that the knock-on proton spectrum contains almost no particles of energy higher than 100 Mev.

It has been shown by Shamov⁹ that fission accompanied by the emission of a definite number of charged particles can be associated with a definite mean excitation energy. Thus for each group of fissions with emission of a definite number of charged particles we can determine the average fissioning nucleus on the assumption that all particles escape from the nucleus before fission occurs. The charge of the fissioning nucleus is obtained directly from the number of visible tracks with a correction for the invisible tracks. The number of neutrons in the fissioning nucleus can be calculated very accurately by means of the evaporation theory from a knowledge of the excitation energy expended for neutron evaporation alone.

In order to clarify the different fission modes of different nuclei we for each group plotted the distribution of fissions with respect to the range

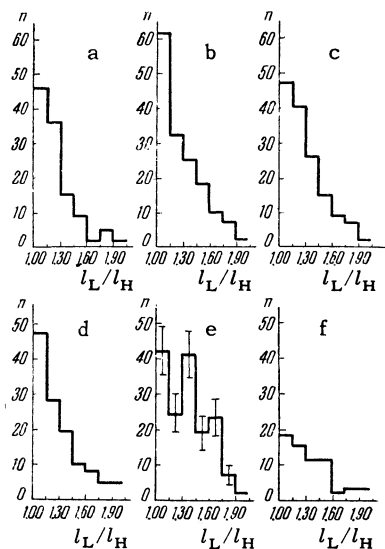


FIG. 1. Distribution of Bi fissions n with respect to the range ratio l_L/l_H of light and heavy fragments from fissions emitting: a-0, b-1, c-2, d-3, e-4 or 5, f-6 or more charged particles. The mean error of measurement of the range ratio is $\Delta(l_L/l_H) = 0.1 l_L/l_H$.

ratio l_L/l_H of light and heavy fragments; the histograms are given in Fig. 1. In the cases of 0, 1, 2, 3 charged particles the highest yield is consistently observed near the ratio $l_L/l_H = 1$, i.e., for symmetric fission. With increasing asymmetry the yields decrease monotonically in all four cases. An entirely different picture results in the case of 4 and 5 emitted charged particles (Fig. 1e): the yield does not decrease monotonically as the asymmetry increases. Unlike the previous relations histogram e shows three maxima, two of which lie far outside the limits of statistical error. A χ^2 test¹⁰ was performed to determine whether statistical fluctuations could account for these departures from the smooth curves for groups a, b, c and d. The area under a smooth curve was set equal to the area of the experimental histogram; then $\chi^2 = 14.4$ for the best fit of the experimental values. For the number of degrees of freedom involved here, $F = 6 - 1 = 5$, the probability of obtaining such statistical fluctuation is $P < 0.02$, which is so small that statistical errors cannot account for the observed curve. We are also not aware of anything that could cause any systematic errors. When 4 or 5 charged particles are emitted there is apparently about the same probability of asymmetric fission into fragments with the given masses as for the familiar symmetric fission.

It is difficult to make any definite statement regarding the mode of fission of nuclei associated with 6 or more emitted charged particles. In the first place, the data are very meager. Secondly,

as was shown in references 11 and 12, Ag and Br stars with 6 or 7 prongs show a very high probability of heavy fragments (including those with range of about 10μ) together with recoiling nuclei, or pairs of heavy fragments, which can be distinguished from Bi fission only with great difficulty. Because of the large AgBr content of emulsions such spurious "fissions" may comprise a considerable fraction. One must therefore exercise great caution before asserting that the fission of a given group is symmetric as shown in Fig. 1f.

The use of nuclear emulsions enables us to include the important combined range of fission fragments in our analysis in addition to the range ratio. While single ranges, because of the extremely great diversity of nuclear products, cannot serve to determine the kinetic energy release involved in fission, the combined range is very closely related to the total kinetic energy of fission fragments. Photographs of fissions provide a measurement of the range ratio of light and heavy nuclei together with the combined range of these fragments. By averaging the combined ranges over a large number of fissions with given asymmetry we can obtain the dependence of the mean combined range $\bar{l} = \overline{l_L + l_H}$ on the ratio l_L/l_H ; this gives approximately the dependence of the total kinetic energy of fragments on the degree of asymmetry. Figure 2 gives $\bar{l} = f(l_L/l_H)$ for two groups of Bi fissions and, for comparison, the same relationship for U fission by thermal neutrons. Unlike the gradually falling curve for U (Fig. 2c), both groups of Bi fissions (Fig. 2a and b) show the same irregular character. For Bi fissions

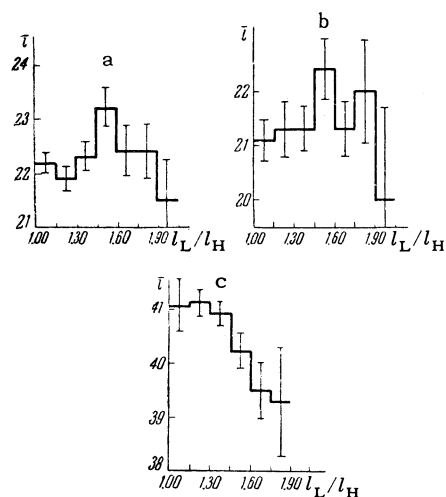


FIG. 2. Mean combined range of fission fragments, $\bar{l} = \overline{l_L + l_H}$, as a function of the range ratio l_L/l_H of light and heavy fragments, for Bi fission accompanied by: a-0, 1, 2, 3; b-4 or 5 charged particles (\bar{l} in μ). c-U fissions by thermal neutrons (\bar{l} in relative units).

accompanied by 0, 1, 2, 3 charged particles the highest value of \bar{l} is reached with range asymmetry 1.52. The mean combined range reaches just as steep a maximum for the same asymmetry in Fig. 2b (for Bi fission with 4 or 5 emitted charged particles). The second maximum in the vicinity of range ratio $l_L/l_H = 1.82$ is not so reliable. The data indicate that the highest value of the total kinetic energy release in Bi fission induced by 660-Mev protons is observed in asymmetric fission.

2. ANALYSIS AND DISCUSSION OF EXPERIMENTAL RESULTS

We used Bohr's relationships in reference 13 in order to relate the range ratio of light and heavy fission fragments to the corresponding charge values. The energy losses of slowed-down heavy fragments are divided into those at velocities higher than v_0 (the electron velocity in the hydrogen atom) and those at velocities lower than v_0 . In the first case ionization loss is dominant, while in the second case nuclear collisions produce most of the energy loss. For the sake of simplicity, Bohr neglects energy losses due to nuclear collisions and extrapolates the expression for ionization loss as far as $v = 0$. For a heavy stopping medium he thus obtains $dv/dl = \text{const}$. The variation of the effective charge as a function of velocity can be represented very well by $Z_1^* = Z_1^{1/3} v/v_0$, whence we easily obtain

$$l_L/l_H = (Z_H/Z_L)^{2/3} f(Z_H, Z_L), \quad (1)$$

where

$$f(Z_H, Z_L) = [3(2Z_H^{1/3})^{-1/3} + (2Z_H^{1/3})^{-1}] / [3(2Z_L^{1/3})^{-1/3} + (2Z_L^{1/3})^{-1}].$$

Since we are using the ratio of the ranges rather than their absolute values constants that are characteristic of the emulsion disappear; this expression contains neither the mass nor the velocity of the fragments. The ratio l_L/l_H depends only on the nuclear charges of the fission fragments. Extrapolation of the expression for ionization loss to small velocities as a substitute for taking nuclear collisions into account when $v < v_0$ does not result in a very large departure from the true value of l_L/l_H . This deviation can be estimated from the data given by Boggild et al,¹⁴ who determined the range-velocity relation for uranium fission induced by thermal neutrons. We find that the value of l_L/l_H obtained from (1) must be increased by about 10%. Thus a practical formula for the charges would be

$$l_L/l_H = 1.1 (Z_H/Z_L)^{2/3} f(Z_H, Z_L^*) \quad (2)$$

The correcting factor is, of course, unnecessary for symmetric fission, i.e., Eq. (1) remains valid near $l_L/l_H = 1$.

Assuming $l_L/l_H = 1.32$ (the experimental value for uranium fission by thermal neutrons), from $Z_H + Z_L = 92$ and (2) we obtain $Z_H = 54$, $Z_L = 38$ in agreement with experiment. Thus (2) is entirely acceptable for the purpose of determining charge asymmetry from the range ratio.

In calculating the charge and mass number of a fissioning nucleus we assume an emission mechanism for Bi fission.⁴ This hypothesis has been supported very convincingly in a number of papers¹⁵ and there are hardly any grounds for doubting the emissive character of Bi fission. The calculation was performed in two steps. (1) Observed charged-particle tracks were used to determine the charge carried from the nucleus by charged knock-on and evaporated particles. The number of fast protons not recorded by the emulsion was estimated from a nuclear cascade calculation.¹⁶ The energy carried away by evaporated charged particles was subtracted from the average excitation energy, which has a definite value for each nuclear group.⁹ The remaining energy is expended only for the evaporation of neutrons. (2) The number of evaporated neutrons was determined by trial and error, with the calculation continuing until the remainder of excitation energy became smaller than the neutron binding energy.* It was thus possible to determine the mean charge and mean mass number of the fissioning nucleus for each group of fissions.

The calculation shows that for groups of fissions accompanied by the emission of 4 or 5 charged particles the average species of fissioning nucleus is Pt_{78}^{174} . The great majority of fissioning nuclei in this group contain less than 100 neutrons. Fissions with the emission of 3 charged particles have an initial excitation energy which is 100 Mev lower and most of the fissioning nuclei contain more than 100 neutrons. The other groups (Fig. 1a, b, c) give fissioning nuclei which are even richer in neutrons.

Applying (2) to the range ratios 1.52 and 1.82, for which in Fig. 2b we observe the maximum kinetic energy release, we obtain 50 and 28, 56 and 22, respectively as the charges of heavy and light fission fragments. For the group with a smaller number of charged particles (Fig. 2a) exactly the same position of the maximum (at $l_L/l_H = 1.52$)

*In the case of nuclei with sufficiently large Z^2/A fission can occur when the excitation energy slightly exceeds the neutron binding energy. There will then be one or two more neutron in the fissioning nucleus than the calculated number, but this does not essentially change the final result.

corresponds to charges 53 and 29 approximately. The charges calculated in this way which are associated with the maximum yields of asymmetric fission (Fig. 1e) are 47 and 31, 53 and 25, for heavy and light fragments, respectively.

We shall now attempt to account for the observed effects. We assume that the shell structure of the product nuclei is the decisive factor that determines the charge and mass distributions of fission fragments. This hypothesis has frequently been advanced by different investigators to account for asymmetric fission.⁷ The symmetric fission which is observed for groups emitting 0, 1, 2, 3 charged particles is then attributed to the influence of the neutron shells of product nuclei. All of the fissioning nuclei of these groups contain more than 100 neutrons and each fragment involved in symmetric fission must contain a little more than 50 neutrons. Thus stable 50-neutron groups induce symmetric deformations of the nucleus resulting in splitting into two identical parts. When a fissioning nucleus is left with less than 100 neutrons (in 4- and 5-pronged fissions) the neutron shells cease to exert their influence while the influence of proton shells containing 50 and 28, or 50 and 20, protons are most influential. Indeed, the mean charge of a fissioning nucleus is $Z = 78$ in this group. Thus when the original nucleus fissions there is a very strong possibility of obtaining product nuclei with 50- and 28-proton shells and a somewhat slighter possibility of obtaining charges somewhat exceeding 50 and 20. The fact that according to the calculation above the maximum-yield fragments (Fig. 1e) have charges 47 and 31 may be attributed either to the estimative character of (2). (Charges 50 and 28, 56 and 22, can possibly correspond to range ratios 1.37 and 1.67.) or to the nature of the actual fission mechanism. In the latter case it can be assumed that asymmetric nuclear deformations are caused by shells of 50 and 28, or 50 and 20 protons. When the critical point has been passed the light fragment in process of formation captures most of the neck between the two portions of the nucleus and receives a greater charge: 31 instead of 28, 25 instead of 20 + n, $n \leq 4$. The symmetric fission peak seen in Fig. 1e evidently results from the most highly excited fissioning nuclei, just as the symmetric uranium fission yield grows as we pass from thermal to faster neutrons.

The dependence of the total kinetic energy of fission fragments on the degree of asymmetry can also be understood to some extent from the given point of view. Although for fission groups with 0, 1, 2, 3 prongs symmetric fission occurs, because

of predominantly symmetric deformations, the highest energy release is observed for fissions into fragments containing about 53 and 29 protons, which are close to the magic numbers 50 and 28. This indicates that with the less frequently encountered types of asymmetric deformations the fragments separate from each other at smaller distances than in the case of symmetric fission.* There is a corresponding increase in the total kinetic energy of the fragments and their excitation energy will be smaller.

For the groups of 4- and 5-pronged fissions the maximum of kinetic energy release is also observed in asymmetric fission. When a nucleus of charge 78 splits into fragments with charges 50 and 28, or 56 and 22, the stable compact product nuclei at the instant of fission are a very short distance apart; this facilitates the greatest release of Coulomb energy.

The foregoing interpretation of our experimental findings is only qualitative and lays claim to neither completeness nor finality. Further investigations are required to improve and supplement the data. We believe that data on fission modes resulting from the bombardment of heavy elements by high-energy particles can be used to answer a number of questions about the fission of weakly-excited nuclei.

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Translated by I. Emin

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