Probabilities of Symmetric and Asymmetric Fission in the Proton Bombardment of Th^{232}†

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The ratio of symmetric to asymmetric fission in the proton bombardment of Th^{232} does not rise steadily with increasing proton energy; a periodic decrease is superposed upon the over-all increase. This is attributed to the changing pattern of various fission reactions, (p,f), (p,nf), etc.

INTRODUCTION

In general, the ratio of symmetric to asymmetric fission increases as the energy applied to the fissioning nucleus increases.[1–15] The number of emitted neutrons associated with fission also increases with increasing energy.[3,14,16] These excess neutrons might be emitted from the nucleus before it undergoes fission,[3,17,18] from the fragments after fission,[14,19] or from the nucleus after it has deformed to cross the fission barrier, but before separation of the fragments.[11] Many competing reactions are possible; for example in proton-induced fission the reactions (p,n), $(p,2n)$, etc.; (p,f); (p,nf), $(p,2nf)$, etc. (where neutron emission precedes fission); (p,fn), $(p,f2n)$, etc. (where fission precedes neutron emission). It is an interesting problem to decide which reactions account for most of the fissions in each particular case of target, projectile, and energy.

It is difficult to distinguish experimentally whether neutron emission precedes or follows fission. We have considered the possibility that studies of the ratio of symmetric to asymmetric fission can provide some information. The occurrence of reactions such as (p,nf), $(p,2nf)$ in which neutron emission precedes fission has the effect of reducing the energy actually present in the fissioning nucleus to some value less than the energy applied by the bombarding particle and this effect should be reflected in the observed value for the ratio of symmetric to asymmetric fission. Fairhall[20] has used this argument in an interesting discussion of his results for the fission of Bi^{209} by 22-Mev deuterons. We report in this paper the results obtained for the proton-induced fission of natural thorium, Th^{232}.

EXPERIMENTAL METHODS

General

The proton-induced fission of Th^{232} has been studied previously by Tewes and James[6] who obtained fission yield curves at several energies and demonstrated the over-all rise in the ratio of symmetric to asymmetric fission with increasing proton energy. We wished to study the change in this ratio with much greater energy resolution and it was expedient to choose for yield measurements two nuclides which would typify the symmetric and asymmetric fission modes rather than measure complete yield curves. The nuclides Ag^{113} and Ba^{139} serve this purpose since according to the results of Tewes and James Ag^{113} lies essentially at the bottom of the trough in the yield curve while Ba^{139} lies essentially at the top of the heavy-mass peak. We are concerned mainly with the proton energy range below about 50 Mev in which range the fission yield curve is "double humped" and the asymmetric modes represented by the peaks are the most probable modes; some measurements were, however, made at energies above 50 Mev. We have also studied the yields of some nuclides other than Ag^{113} and Ba^{139} at a few selected energies and by observing that these yields fall into the expected pattern we confirm to some extent the reliability of our techniques, in particular of our counting methods, and also that Ag^{113} and Ba^{139} do in fact lie at the trough and peak of the yield curve respectively.

Irradiations

The targets consisted of strips of 0.004-in. aluminum foil coated on the front surface with ThO_2 of thickness

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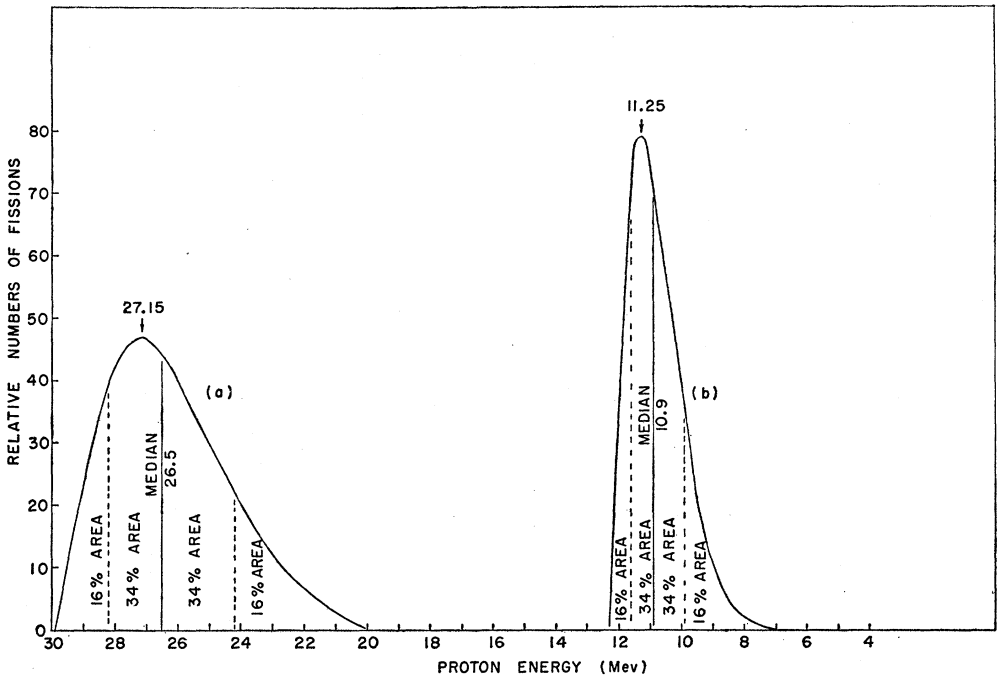

FIG. 1. Relative numbers of fissions occurring at various proton energies for target radii corresponding to maximum proton energies of 30 Mev (curve *a*) and 12.5 Mev (curve *b*).

1.0 mg/cm^2 and wrapped in one layer of 0.001-in. aluminum foil. Experiments at low energies (6.3–9.5 Mev) were made with targets of thorium metal foil, thickness 0.0015 in. The measurements with ThO_2 targets were carried down to 8.5 Mev so that results for both types of targets were available in the energy range 8.5–9.5 Mev; these were found to be in agreement. Behind the target was a $\frac{1}{2}$-in. thick block of aluminum which served to prevent multiple traversals of the target by the beam. The yield of fission products is very small at low proton energies and it seemed advisable to establish that the observed fissions were due only to protons and not to γ rays or neutrons originating in the target holder or elsewhere in the cyclotron tank. A number of experiments were made on the distribution of fission products across the foils, i.e., along the cyclotron radius; proton-induced fissions are localized in a narrow band at the edge of the target foil whereas γ-ray or neutron fissions should be more uniformly distributed across the foil. Another series of experiments was made using targets covered with sufficient aluminum to stop the proton beam. The results showed that γ-ray and neutron fissions were below the limit of detection. We were also able to show that no pertinent activities were induced in the target backing foil or covering foils in the absence of target material. Irradiation times varied from 1 minute to 1 hour.

Proton Energy

The proton beam inside the Harwell cyclotron has a current of about 1 microampere. The current was not measured since only relative fission yields were required. The proton energy defined by the target's radial position is the maximum energy of the protons striking the target. There is a spread of energy below this maximum due to precession of the center of rotation of the beam. To obtain the energy effective in producing fissions the following corrections were applied: (1) for the actual energy distribution of the protons,[21] (2) for the energy lost by the protons in passing through the 0.001-in. aluminum covering foil and half-way through the target of oxide or metal foil,[22] and (3) for the variation of fission cross section with proton energy.[23,24]

Corrections 1 and 2 give the relative numbers of protons of each energy in the target material for a given target radius and if this is multiplied by the appropriate cross section a graph can be constructed showing the relative number of fissions occurring at each energy. Figures 1(a) and 1(b) show examples calculated for target radii corresponding to 12.5- and 30.0-Mev maximum proton energies. Curves were constructed for representative target radii over the required range. The curves were integrated graphically and the energy quoted in our results is the median energy, i.e., that corresponding to half area. The energy spread indicated is that corresponding to 34% area on either side of the median, i.e., 68% of the fission occurred within the energy range indicated.

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Separation of Fission Products

The fission products were isolated and purified by standard radiochemical methods using carrier quantities in the order of 10 mg per element. The whole of the target foil and covering foil were dissolved in the carrier-containing solution, thus ensuring that all the fission fragments were obtained without any differentiation between species which might have arisen from their different ranges. The silver was first separated from other materials 15–30 minutes after the end of irradiation, with hold-back carriers being used for cesium, barium, palladium, and other elements when required. This silver was then allowed to stand for 2 hours before purification; this allowed decay of the Ag^{115} (21 min) to Cd^{115} before purification from cadmium for which hold-back carrier was used. The barium was separated 90 min after irradiation, by which time Cs^{139} (9.5 min) had completely decayed to Ba^{139}. The final precipitates were collected on small filter-paper disks and weighed to obtain the chemical yield.

β-Counting Methods

The precipitates were counted in a methane-flow proportional β counter of approximately 2π geometry and having an absorber of 5.3-mg/cm^2 aluminum over the sample. The counting efficiency of this arrangement for the fission products was known from the measurements of Cuninghame *et al.*,[25] who compared the observed activities at various precipitate weights and with various absorbers to the disintegration rates measured by 4π β counting. In the case of the Ag^{113} and Ba^{139} measurements, the precipitates were of nearly the same weight in every experiment and thus the counting correction was nearly constant throughout. The actual "counting" was done by an automatic sample-changer coupled with automatic scaling, timing, and printing devices.

Calculation of Results

The usual decay curves were constructed and the long-lived components, whose contributions were small, were subtracted. The half-lives obtained agreed with accepted values, in particular Ag^{113}, 318 min and Ba^{139}, 85 min, except that the Ag^{113} samples obtained with proton energies above 30 Mev showed half-lives shorter than this figure. This is because at high energies Ag^{112} (3.1 hr) has a significant independent yield, whereas at low energies Ag^{112} is formed only via Pd^{112} which is comparatively long-lived (21 hr) and permits only a very small growth of Ag^{112} under our experimental conditions. From the observed half-life it was possible to make a rough estimate of the extent of the Ag^{112} contamination in the Ag^{113} samples. Figure 2 shows how this varies with proton energy; the solid line shows the Ag^{112} activity as a percentage of the Ag^{113} activity and the dashed line shows the independent fission yield of Ag^{112} as a percentage of the cumulative Ag^{113} yield calculated with the assumption that the β-counting efficiency was the same for both nuclides. The contamination of about 2% at energies below 30 Mev is accounted for by the small growth of Ag^{112} via Pd^{112}. The Ag^{113} results were corrected by using the data of Fig. 2.

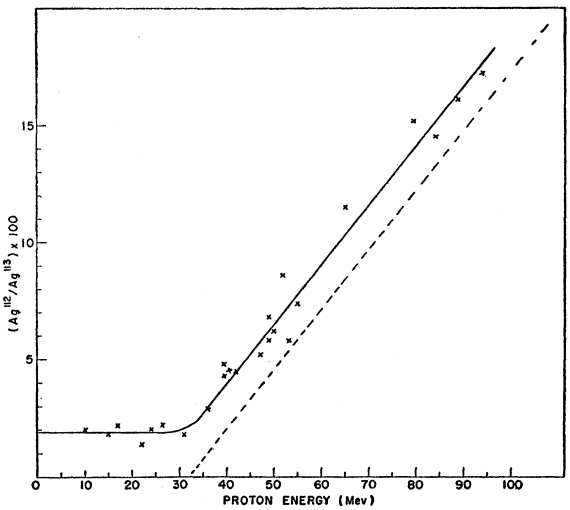

FIG. 2. Ag^{112}—Ag^{113} ratio *vs* proton energy. Solid line: Ratio of activities 260 min after end of 5-min proton bombardment; first Ag separation 30 min after end of bombardment. Dashed line: Ratio of independent yield of Ag^{112} to cumulative yield of Ag^{113}.

The relative fission yields were calculated under the assumptions that, in the first place, Pd^{113} has such a short half-life (1.5 min) that the Ag^{113} can be regarded as formed directly in fission and secondly that all the Ba^{139} was formed via Cs^{139} (9.5 min). The first assumption introduces an error of less than 0.5% in the results. The second assumption is probably not completely valid since Ba^{139} presumably has some independent yield at the higher energies. If we accept the Ag^{112} case as analogous, we find that even at 100 Mev about 80% of the Ba^{139} is formed via Cs^{139}. The error in the results from the second assumption should not be greater than about 2% at 100 Mev and should be less at lower energies. Activities were obtained by calculation using all the available counting data rather than by graphical methods since this former procedure gave better precision in the results.

RESULTS AND DISCUSSION

Measurements of the Ag^{113}/Ba^{139} ratio at 58 different proton energies were made. At 19 of these energies duplicate measurements were made and at 3 energies triplicate measurements. From the results of the repeated measurements the precision of the results appears to be about $\pm 2\%$, and from considering the various sources of error we estimate the accuracy of the results to be about $\pm 5\%$.

[25] Cuninghame, Sizeland, and Willis (private communication).

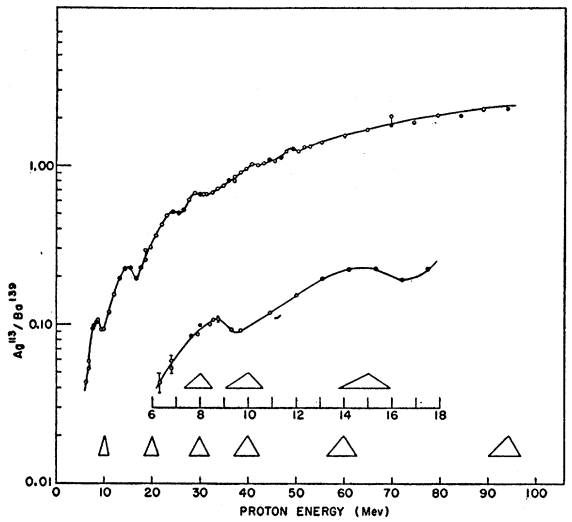

FIG. 3. Ratio of $Ag^{113}-Ba^{139}$ yields *vs* proton energy. Notes: (1) The size of the circles shows the ±2% statistical error. (2) The solid circles show points which upon repetition were reproduced within the ±2% statistical error. (3) Points which upon repetition did not coincide within ±2% are shown as open circles separated vertically. (4) The spread in energy of the protons is indicated by open triangles.

The results are expressed graphically in Fig. 3. Superimposed upon the over-all rise of the valley/peak ratio with increasing energy are a number of dips, rather pronounced in the lower energy regions but becoming less pronounced at higher energies. These dips are located at proton energies of 9.5, 16.4, 25.5, 31, 42–45, 49–52 Mev. The position and indeed the existence of the two highest energy dips is somewhat speculative, but the four low-energy ones are quite clearly defined. The effect would probably have been more pronounced had the spread in proton energies been less.

The Ag^{113} and Ba^{139} yields observed at high energies do not measure the total yields of the mass 113 and 139 chains since at energies above about 50 Mev the independent yields of stable nuclei become significant. Furthermore, above this energy the Ag^{113} yield is greater than the Ba^{139} yield, showing that the fission yield curve has lost its double-humped character and the concept of the peak/valley ratio is no longer applicable. We have therefore made comparatively few measurements in the energy range 50–100 Mev, sufficient only to indicate that symmetric fission predominates and that the $Ag^{113}-Ba^{139}$ ratio appears to approach a limiting value asymptotically at high energy.

We interpret the results as follows. The excitation energy of the compound nucleus Pa^{233} is the kinetic energy of the proton plus 5.51 Mev, the proton binding energy. While the excitation energy is in the range 6–14 Mev the principal reactions will be (p,f) and (p,n); the energy of the fissioning nucleus Pa^{233} increases with increasing proton energy and the $Ag^{113}-Ba^{139}$ ratio increases likewise (most of this energy range is not accessible experimentally because of the Coulomb barrier). At an excitation energy of approximately 14 Mev the reaction (p,nf) becomes possible, the energy required for this process being composed of the neutron binding energy (6.74 Mev), the kinetic energy of the emitted neutron (about 2 Mev), and the energy required to cause fission in the residual Pa^{232} nucleus (5–6 Mev). The fissions in Pa^{232} [the (p,nf) reaction] occur with an excitation energy approximately 8.74 Mev lower than those in Pa^{233} [(p,f) reaction] and the $Ag^{113}-Ba^{139}$ ratio is lower. The observed $Ag^{113}-Ba^{139}$ ratio, corresponding to a mixture of the two fission reactions, thus falls at around this energy. A further increase in proton energy causes both the (p,f) and (p,nf) reaction to occur with higher energies and the $Ag^{113}-Ba^{139}$ ratio thus rises again until the $(p,2nf)$ reaction sets in, whereupon another fall occurs, and so on.

It is not possible to calculate the precise energy at which the dips should occur because we do not know, in the (p,nf) reaction for example, exactly how much energy must remain in the residual nucleus after neutron evaporation in order to make this reaction have a probability comparable with that of the (p,f) reaction. However, it should be possible to estimate the energy spacing between dips fairly accurately from a consideration of neutron binding energies. We have calculated the spacing and "normalized" the calculations and experimental results by arranging the calculated position of the second dip to coincide with the observed dip at 16.4 Mev. The positions are calculated for proton energies rather than excitation energies. The binding energy of the proton to the Th^{232} target nucleus (5.51 Mev) was taken from the compilation of Glass, Thompson, and Seaborg.[26] The neutron binding energies in the various Pa nuclei were obtained from the same compilation. The kinetic energy of the evaporated neutrons was taken as 2.0 Mev.[27,28] This latter quantity might increase with increasing bombarding proton energy; thus Heckrotte[28] has calculated that for U^{235} with 50-Mev protons the average energy of the evaporated neutrons is 2.05 Mev while with 100-Mev protons

TABLE I. Calculated and observed positions for minima in Fig. 3

Isotope	E_n (Mev)[a]	Incident proton energy at position of minima (Mev) Calc	Observed
Pa^{233}	8.74	8.9	9.5±0.5
Pa^{232}	7.54	16.4	16.4±0.5
Pa^{231}	8.76	25.2	25.5±0.5
Pa^{230}	7.73	32.9	31.0±1.0
Pa^{229}	9.14	42.1	43.5±1.5
Pa^{228}	8.08	50.2	50.5±1.5

[a] E_n = energy to evaporate one neutron = neutron binding energy + 2 Mev.

[26] Glass, Thompson, and Seaborg, J. Inorg. Nuc. Chem. **1**, 3 (1955).
[27] D. Skyrme (private communication).
[28] W. Heckrotte, University of California Radiation Laboratory Report UCRL-2184 (unpublished).

it is 3.04 Mev. This effect would increase the spacing between dips at higher energies. Table I shows the comparison between calculated and observed positions.

Confining our attention to the first three dips, we note that they take place within a rather narrow energy range, 1–2 Mev, after which the curve rapidly resumes its upward trend with a slope not greatly different from that preceding the dip. We interpret this as showing that each reaction (p,nf), $(p,2nf)$, $(p,3nf)$, $\cdots$ sets in rapidly and that thereafter the competition does not change greatly until a new reaction becomes possible. See also Fig. 4.

We can make a rough quantitative estimate of the contributions of each fission reaction in the following way. Consider the situation when the (p,nf) reaction has just set in, i.e., at the bottom of the first dip. Let σ_0 be the proportion of (p,f) fissions and σ_1 the proportion of (p,nf) fissions; let Ag_0 and Ba_0 be the yields from (p,f) fission and Ag_1 and Ba_1 those from (p,nf) fission. The observed Ag—Ba ratio, R, is given by

$$R=(\sigma_0 Ag_0+\sigma_1 Ag_1)/(\sigma_0 Ba_0+\sigma_1 Ba_1).$$

The (p,nf) reactions are occurring at an energy not much above threshold and therefore Ag_1 is very small. The Ba yields, being on the peak of the yield curve, do not change very much with energy and hence $Ba_0 \approx Ba_1$. Hence:

$$R\approx\sigma_0 Ag_0/[(\sigma_0+\sigma_1)Ba_0].$$

The ratio Ag_0-Ba_0 is the ratio applicable to the (p,f) reaction uncontaminated by the (p,nf) reaction and can be estimated by extrapolating the portion of the curve prior to the dip. The extrapolation is more easily done if the experimental data is plotted with (proton energy)$^{-\frac{1}{2}}$ as abscissa because the interdip portions thus become straight lines, Fig. 4 (see references 6, 7, 12, 13). If r is the Ag—Ba ratio obtained by such an extrapolation

$$R=\sigma_0 r/(\sigma_0+\sigma_1) \quad \text{and} \quad \sigma_1/\sigma_0=(r-R)/R.$$

Applying the same treatment at the second dip we obtain the ratio of the $(p,2nf)$ fissions, σ_2, to the sum of the (p,f) plus (p,nf) fissions, $\sigma_0+\sigma_1$. From the third dip we obtain the ratio of $(p,3nf)$ fissions, σ_3, to the sum of (p,f) plus (p,nf) plus $(p,2nf)$ fissions, $\sigma_0+\sigma_1+\sigma_2$, and so on. Table II shows the values obtained from

TABLE II. Proportion of fissions due to the reaction (p,xnf) for various values of x.[a]

R	r	$\left(\frac{\sigma_x}{\sigma_0+\sigma_1\cdots\sigma_{x-1}}\right)$	x
0.090	0.17	0.90	1
0.20	0.33	0.65	2
0.50	0.66	0.32	3
0.66	0.82	0.24	4
1.1	1.3	0.18	5
1.4	1.6	0.14	6

[a] See text for explanation of symbols.

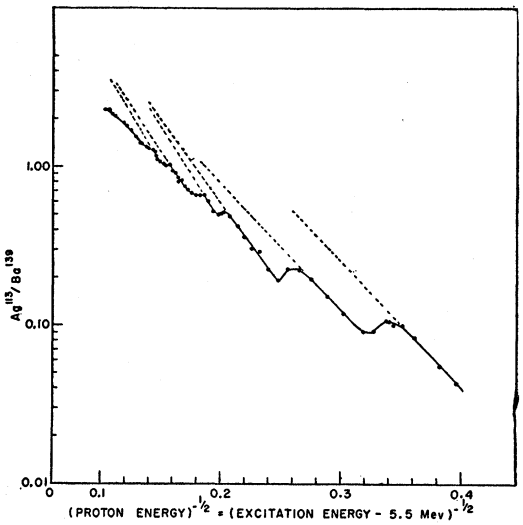

FIG. 4. Ratio of $Ag^{113}-Ba^{139}$ yields *vs* (proton energy)$^{-\frac{1}{2}}$.

such an analysis. They are necessarily quite approximate due to the assumptions made and to the energy spread in the protons used in the experiment, but they justify the following qualitative conclusions. The value of $\sigma_x/(\sigma_0+\sigma_1\cdots\sigma_{x-1})$ falls as x increases, which is reasonable since in order for the reaction (p,xnf) to occur the excited compound nucleus must survive against all the other fission reactions which are possible during the earlier stages of the neutron evaporation. Since the value does not fall rapidly, the chances of survival must be considerable, at least as far as $(p,4nf)$ and probably further. Thus fission competes poorly against neutron emission in the Pa nuclei. The value of 0.9 for the relative probabilities of the reactions $Th^{232}(p,nf)$ and $Th^{232}(p,f)$ with protons of $\sim$10 Mev can be compared with the value of 1.3 for the relative probabilities of the reactions $Th^{232}(n,nf)$ and $Th^{232}(n,f)$ with neutrons of $\sim$10 Mev obtained from fast-neutron fission cross-section data.[29]

The fact that the dips in Fig. 3 disappear at higher proton energies is due partly to the greater energy spread of the higher energy protons but probably has more fundamental causes in addition. In the first place at high energies there will be overlapping of reactions such as $(p,4nf)$, $(p,5nf)$, $(p,6nf)$, etc., because the energy carried away by the evaporated neutrons is variable. Secondly, as mentioned above, reactions of the type (p,xnf) are expected to become improbable at high values of x since to undergo such reactions the nucleus must survive against fission which can occur earlier during the neutron boil-off process. During the boil-off process the nucleus is becoming more fissionable while the neutron binding energy is increasing. How high x must be before reactions (p,xnf) become insignificant will depend upon the fissionability of the target

[29] J. D. Jackson, Proceedings of the Symposium on the Physics of Fission, held at Chalk River, May 14–18, 1956, CPP-642-A (unpublished), pp. 125–139.

nucleus. If the target is very fissionable it is to be expected that fission will compete more effectively against neutron emission during the early stages of boil-off and thus contributions from (p,xnf) reactions will disappear more quickly with increasing values of x than is the case in less fissionable species. Glass *et al.*[14] have concluded that in the reaction of Pu^{239} with helium ions (compound nucleus Cm^{243}) the chain of successive neutron emission is very quickly interrupted by competition from fission and that the excess neutrons must therefore be emitted from the fission fragments.

Comparatively small changes in the nature of the compound nucleus might well result in large changes in the pattern of reactions. For example, in the simple case of the competitive reactions (γ,f) and (γ,n) with γ rays of 17–20 Mev, the probability of fission is 6% for Th^{232} and 60% for Pu^{239}.[30] Such differences would be magnified in cases where a succession of competitive reactions occurred. We are therefore extending our measurements to other target nuclei.

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[30] Huizenga, Grindler, and Duffield, Phys. Rev. **95**, 1009 (1954).

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Coulomb Effects in Inner Bremsstrahlen*

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The Coulomb correction to the photon spectrum accompanying beta decay is calculated, treating the Coulomb field as a perturbation. It is shown that for allowed and unique first forbidden transitions, the result differs from that of Knipp-Uhlenbeck-Bloch only by the appearance of an extra factor, related to the Sommerfeld factor in ordinary bremsstrahlen. Analytic formulas are presented for these two selection rules, and a comparison made with recent experiments.

I. INTRODUCTION

THE original calculations[1] of the intensity of the photon spectrum accompanying beta decay have long been successful in explaining the observed data,[2] in spite of the expected inaccuracy due to the use of plane wave functions rather than Coulomb wave functions for the electrons. Recently, the first deviations from these predictions have been reported[3] in the spectrum of photons emitted by P^{32}, S^{35}, and Y^{90}. It is the purpose of this note to report the result of a derivation of the correction to the photon spectrum due to the Coulomb field of the nucleus, and to compare this result with these experiments. The Coulomb field is treated using perturbation theory; that is, only these additional terms proportional to Z are retained. Both allowed and forbidden transitions will be treated.

A few words in justification of this method of calculation are appropriate. It is not immediately obvious why the original calculation is so successful, or why additional terms in the perturbation theory would be expected to adequately treat the effect of the Coulomb field. One might expect for example, that a first order calculation would add terms of order $Z/137$, which is not a small correction in moderately heavy nuclei. In fact, our results indicate that the necessary corrections to the KUB formula are generally smaller than this. The fundamental reason for the accuracy of the KUB formula is that it has been used to predict only the photon intensity *relative to the beta intensity*, and not the absolute photon intensity. The number of photons per beta decay is a quantity which is comparatively independent of the atomic number, owing to a partial cancellation of the Coulomb effects on the photon and beta intensities. Thus, it is hoped that if we calculate this quantity to first order in Z, a similar partial cancellation of the higher ordered terms will considerably extend the validity of the perturbation theory, which would otherwise be expected to be valid only for light nuclei and high-energy decays.

As an example of this cancellation, we can consider the effect of attempting to correct for the Coulomb field

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[2] For a recent review see C. S. Wu, in *Beta and Gamma Ray Spectroscopy* (Interscience Publishers, New York, 1955).
[3] K. Liden and N. Starfelt, Phys. Rev. **97**, 419 (1955); N. Starfelt and N. L. Svantesson, Phys. Rev. **97**, 708 (1955); H. Langevin-Joliot, Compt. rend. **241**, 872 (1955); M. Goodrich (private communication).