Au^{198} beta particles were computed according to Eq. (10). The results are shown in Table I. The geometrical corrections for the finite sizes of the gamma detector and of the scattering foil were taken into account on the basis of the Θ dependence given by Eqs. (5) and (6).

4. DISCUSSION

In the ξ approximation the degree of transverse polarization $P_T(\Theta,W)$ of the beta particles in a beta-gamma correlation experiment is related to the anisotropy factor $A_2(W)$ of the corresponding beta-gamma directional correlation according to Eqs. (5) and (6).

The present measurements of the transverse polarization were made at an average beta energy of $\bar{W}=2.0$ mc^2, for which one takes from reference 8, $A_2(\bar{W}=2.0)=+0.018\pm0.001$. With this value one obtains for the degree of transverse polarization in the beta-gamma plane:

$$P_{T\parallel}(\Theta=135°, \bar{W}=2.0)=+0.006,$$

and for the polarization perpendicular to the beta-gamma plane:

$$P_{T\perp}(\Theta=135°, \bar{W}=2.0)=-0.003.$$

Within limits of error the experimental values of P_T agree satisfactorily with the values predicted by the ξ approximation for first-forbidden nonunique beta transition.

ACKNOWLEDGMENTS

The authors are indebted to J. E. Alberghini for his assistance in some of the measurements and to Mrs. M. Kawaguchi for her help in reading and processing the large amount of automatically accumulated data. The authors also would like to express their appreciation to F. Long for his cooperation in building the vacuum chamber. One of the authors (P.C.S.) is grateful to the Eastman Kodak Company for the grant of a fellowship.

PHYSICAL REVIEW VOLUME 118, NUMBER 3 MAY 1, 1960

Fission of Ra^{226} by Deuterons and Helium Ions*†

R. C. JENSEN‡ AND A. W. FAIRHALL
Departments of Chemistry and Physics, University of Washington, Seattle, Washington
(Received November 30, 1959)

Fission induced in Ra^{226} by 14.5- and 21.5-Mev deuterons, and by 23.5-, 31-, and 43-Mev He ions has been studied using radiochemical techniques. The mass distributions of fission products for deuteron-induced fission is triple-humped, corresponding to separate symmetric and asymmetric fission modes. The symmetric mode dominates at the higher bombarding energy. The mass distributions observed for fission products from He-ion induced fission look more "normal": asymmetric at the lowest bombarding energy, becoming a single broad peak at the highest bombarding energy. These results are interpreted in terms of a symmetric fission mode which increases strongly with increasing excitation energy, and an asymmetric fission mode which occurs mainly at low excitation energies following neutron evaporation from highly excited compound nuclei. Asymmetric fission is interpreted to be disappearing as a fission mode for nuclei of lower atomic number than thorium.

I. INTRODUCTION

In a previous paper[1] we reported the mass distribution of fission fragments from fission induced in Ra^{226} by 11-Mev protons. In those experiments, where radiochemical techniques were employed, the mass distribution of fission fragments was found to be a novel one: the mass-yield curve was triple-humped, corresponding to separate symmetric and asymmetric fission modes. Indications of a similar result have been reported[2] for neutron-induced fission of Ra^{226}, where counter techniques were employed: at low neutron energies the distribution of fragment kinetic energies falls into two groups, indicating asymmetric fission. At neutron bombarding energies around 15 Mev a prominent single peak is observed for the fragment kinetic energies, corresponding to symmetric fission. At intermediate neutron bombarding energies the fragment kinetic energy distribution indicates a transition from asymmetric to symmetric mass division with increasing bombarding energy. Along with the changing character of the mass distribution the cross section for fission was observed to increase sharply with neutron bombarding energy.

Preliminary results of fission incuded in Ra^{226} by 23-Mev bremsstrahlung[3] indicate that the mass-yield

* This work was supported in part by the U. S. Atomic Energy Commission.

† Part of a thesis submitted by R. C. J. in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

‡ Now in the Department of Chemistry, San Diego State College, San Diego, California.

[1] R. C. Jensen and A. W. Fairhall, Phys. Rev. 109, 942 (1958).

[2] R. A. Nobles and R. B. Leachman, Nuclear Phys. 5, 211 (1958).

[3] R. B. Duffield, R. A. Schmitt, and R. A. Sharp, *Proceedings of the Second International Conference on Peaceful Uses of Atomic Energy, Geneva*, 1958 (United Nations, Geneva, 1958), Vol. 15, p. 202, Paper 678.

curve of fission fragments is triple-humped, very similar to the corresponding curve for 11-Mev proton-induced fission.

These results imply that the fission behavior of species of atomic numbers 88 and 89 is different from the heavier elements, ($Z \geqq 90$), and further experiments on fission induced in radium targets are in order. The present paper summarizes the results of experiments on fission induced in Ra^{226} by deuterons and helium ions of several bombarding energies.

II. EXPERIMENTAL PROCEDURES

The experimental procedures used in the experiments being reported here are essentially the same as those described previously for fission studies with protons.[1] The radium was always bombarded in the form of a thin deposit of radium carbonate sandwiches between thin (2.7 mg/cm^2) gold foils. This sandwich in turn was placed between sheets of aluminum foil which served to catch fission fragments escaping from the gold-radium target. Following the bombardment the aluminum catcher foils were processed radiochemically for the desired fission products, which were then counted as described previously. From the counting data, chemical yields, etc., relative fission yields were computed, and referred, as before, to Ag^{111} which was separated in every experiment.

Owing to the finite source thickness fragments which travel at oblique angles relative to the target sandwich normal do not escape. Because of differences in ranges of light vs heavy fragments fewer heavy fragments (by about 20%) escape than complementary light fragments. Correction factors were applied to correct for this effect assuming isotropic emission of fragments. This assumption is probably essentially correct for the case of proton-induced fission, but is not correct for deuteron- and helium-ion induced fission where significant anisotropies have been observed.[4] However, the observed anisotropy is small in the case of deuteron-induced fission and correction factors applied on the assumption of isotropy are probably not seriously in error. In the helium ion case, where anisotropies are larger, the correction factors are probably too large by perhaps 10% for heavy asymmetric fragments, but in any case, as we shall see, the results are sufficiently clear-cut that these corrections do not obscure the significance of the experimental results.

TABLE I. Relative yields of fission fragments from radium bombarded with deuterons and He ions of various energies.

Fission product	Relative fission yield (arbitrary units) 21.5-Mev deuterons	14.5-Mev deuterons	43-Mev He ions	31-Mev He ions	23.5-Mev He ions
Br^{83}	1.4±0.1	2.4	...	...	...
Sr^{91}	2.2±0.2	3.1±0.2	5.2±0.4	6.6	10.8
Sr^{92} [a]	2.1±0.1	2.3	4.4±0.2	5.6	8.6
Sr^{92} [b]	2.6	1.9	4.4	4.1	...
Y^{92}	2.2±0.5	3.0±0.4	3.8±0.4	5.7	...
Y^{93}	2.8±0.2	3.9±0.4	5.2±0.2	5.0	...
Zr^{97}	2.7±0.2	2.2±0.2	4.2±0.2	3.7	...
Mo^{99}	3.8±0.2	3.0±0.2	5.4±0.2	4.3	5.0
Ru^{105}	4.5±0.2	4.5±0.2	5.2	4.1	...
Pd^{109}	4.4±0.2	4.7±0.2	5.4	4.7	4.6
Pd^{112} [c]	5.2±0.1	5.3±0.2	5.6	5.1	4.0
Pd^{112} [d]	5.6±0.2	5.9±0.3	5.6±0.2	4.5	4.4
Ag^{111} [e]	5.0	5.0	5.0	5.0	5.0
Ag^{113}	4.0±0.1	3.9±0.1	4.2±0.2	3.9	3.9
Cd^{115}	4.7±0.1	4.6±0.2	5.0	4.5	4.4
Sb^{127} [f]	...	1.9	...	...	...
I^{131}	2.6±0.4	3.3±0.2	4.0	...	...
I^{133}	3.3±0.4	5.5±0.2	5.6	...	...
Ba^{140} [g]	2.1±0.3	2.4	3.7±0.5	7.3	11.7

[a] By analysis of the total decay curve for Sr^{91} and Sr^{92}.
[b] By milking the Y^{92} daughter from Sr^{92}.
[c] By analysis of the total decay curve for Pd^{109} and Pd^{112}.
[d] By milking the Ag^{112} daughter from Pd^{112}.
[e] The fission yields of all other fission products are measured relative to this nuclide.
[f] By milking the Te^{127} daughter from Sb^{127}.
[g] By milking the La^{140} daughter from Ba^{140}.

Some of the experiments to be reported were performed at the full energy of the deuteron or helium ion

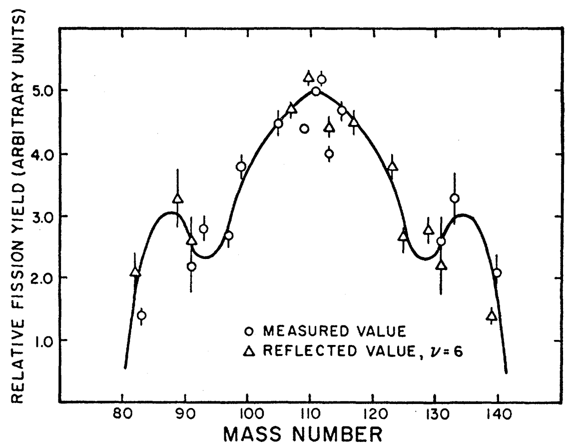

FIG. 1. Mass distribution of fission fragments from fission induced in Ra^{226} by 21.5-Mev deuterons. The initial compound nucleus is Ac^{228} excited to about 29.3 Mev.

beam where the energy of the bombarding particle is known to within a few tenths of a Mev. For other experiments at lower bombarding energies the primary beam was degraded with stainless steel absorbers to the desired energy. In some cases this energy was close to the coulomb barrier for the bombarding particle, so that small variations in the particle would be expected to result in a significant change in the penetrability of the particle into the nucleus. This leads to considerable uncertainty in the calculation of the fraction of the total reaction cross section at a given bombarding energy which goes into fission.

III. EXPERIMENTAL RESULTS

Relative fission yields for a number of selected fission products have been measured for Ra^{226} bombarded with 21.5-Mev and 14.5-Mev deuterons and for 43-Mev, 31-Mev, and 23.5-Mev helium ions. The data are summarized in Table I. Except for 23.5- and 31-Mev helium

[4] C. T. Coffin and I. Halpern, Phys. Rev. 112, 536 (1958).

ion-induced fission, where each nuclide was measured only once, most of the data are the result of several measurements. For these cases the quoted errors are the standard deviations of the measured values. In some instances a particular fission product could be measured either directly or indirectly *via* a daughter activity. The extent to which the two methods agree gives an indication of the reliability of the several

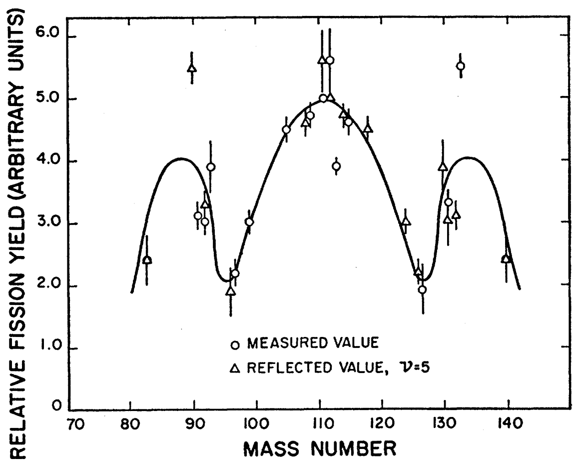

FIG. 2. Mass distribution of fission fragments from fission induced in Ra^{226} by 14.5-Mev deuterons. The initial compound nucleus is Ac^{228} excited to about 22.3 Mev.

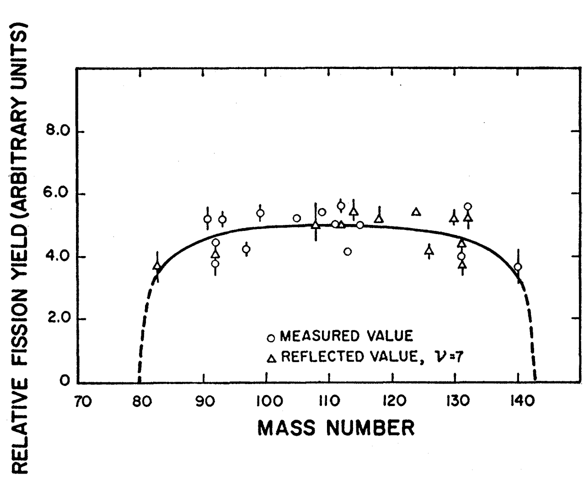

FIG. 3. Mass distribution of fission fragments from fission induced in Ra^{226} by 43-Mev He ions. The initial compound nucleus is Th^{230} excited to about 38 Mev.

correction factors which were applied to counting data to correct for absorption and scattering effects.

The relative fission yields of Table I are plotted vs mass numbers in Figs. 1 to 5. The yields of complementary masses are plotted as reflected points assuming formation of a compound nucleus and the emission of 4, 5, or 6 neutrons during the fission process. The data are not critically dependent on the particular value of ν which is chosen, and the values shown in the figures

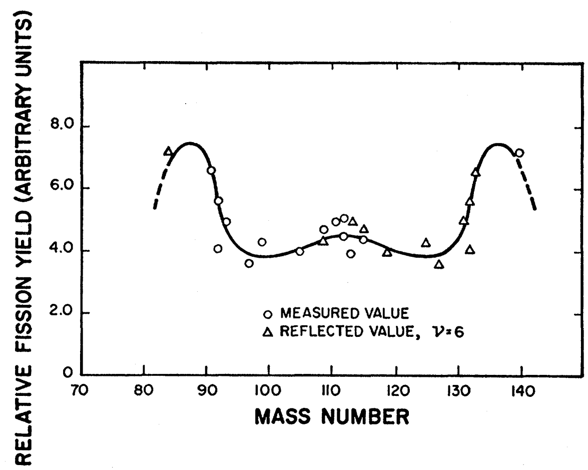

FIG. 4. Mass distribution of fission fragments from fission induced in Ra^{226} by 31-Mev He ions. The initial compound nucleus is Th^{230} excited to about 26 Mev.

represent reasonable values from the point of view of the excitation energies of the initial compound nuclei and the usual number of 2 or 3 post-fission neutrons.

While there is considerable scatter in much of the data, one nuclide, 5.3-hr Ag^{113} appears to have a consistently low fission yield relative to neighboring species. We attribute this effect to independent formation and β decay of isomeric 1.2-min Ag^{113m}, a species which we could not observe in these experiments. The yield of I^{133} appears to be too high in the bombardment with 14.5-Mev deuterons. While duplicate measurements agreed rather closely, some undetermined experimental error is probably responsible for the abnormally high value in this case.

IV. CROSS SECTIONS FOR FISSION

From a knowledge of such things as the amount of radium in the target, the beam current incident upon

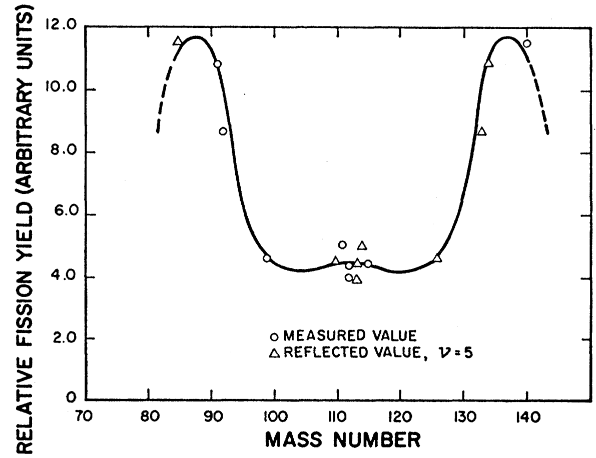

FIG. 5. Mass distribution of fission fragments from fission induced in Ra^{226} by 23.5-Mev He ions. The initial compound nucleus is Th^{230} excited to about 19 Mev.

TABLE II. Summary of fission and total reaction cross sections for bombardment of Ra^{226} by various particles.

Projectile	Compound nucleus	Initial excitation energy (Mev)	Formation cross section Ag^{111}(mb)	Normalized fission yield Ag^{111}(%)	Fission cross section (mb)	Total reaction cross section (mb)	Fission branching ratio (%)
21.5-Mev *d*	Ac^{228}	29.3	2	5.0	80	1700	4.7
14.5-Mev *d*	Ac^{228}	22.3	0.27	4.5	12	800	1.5
43-Mev He^4	Th^{230}	38.2	5.5	3.6	310	1450	21
31-Mev He^4	Th^{230}	26.2	1.5	3.0	100	770	13
23.5-Mev He^4	Th^{230}	18.7	0.1	2.1	10	195	5
10.5-Mev *p*	Ac^{227}	15.5			2 ± 1[a]	120	1.7 ± 0.8

[a] See reference 1.

it, and the disintegration rate of some particular fission product, corrected for counting efficiency, chemical yield, etc, it is possible to compute the formation cross section for that fission product at a particular bombarding energy and for a particular bombarding particle. If the absolute fission yield of the particular fission product is known also, the total fission cross section may be computed. In Table II we have attempted to compute total fission cross sections for the present experiments. The experimental data were used to calculate the formation cross sections for the fission product Ag^{111}. Absolute fission yields were estimated from normalizations of the mass-yield curves passed through the data of Figs. 1–5. Clearly, from the scatter in the data the normalization is somewhat arbitrary, particularly in the case of deuteron-induced fission of Ra^{226}. In drawing the "best" curves through the data of Figs. 1–5 it was assumed that where asymmetric fission appeared to stand out by itself the distribution of asymmetric fragment masses was similar to that which was observed for the asymmetric fission component of 11-Mev proton-induced fission. The latter, in turn, is similar to asymmetric mass yields from the heaviest elements in general. Another characteristic which appears to be true in general for asymmetric fission is that the low mass side of the light fragment "wing" drops sharply below mass 80. This served as a guide in estimating yields of very asymmetric fragments in Figs. 3, 4, and 5.

In view of the many uncertainties in the data the fission cross sections tabulated in Table II are probably accurate only within a factor of about 2.

Also included in Table II are theoretical values[5,6] for the total reaction cross sections for the various bombarding particles and bombarding energies which were used. These values are useful in estimating the fraction of the total cross section which is fission.

Table II also lists the estimated fission and total cross sections for 11-Mev proton-induced fission[1] of Ra^{226}. Because the total reaction cross section is changing rather rapidly for proton bombarding energies near 11 Mev there is considerable uncertainty in the total reaction cross section owing to uncertainties in the bombarding energy. A bombarding energy of 10.5 Mev was used in computing the reaction cross section in this particular case.

V. DISCUSSION

The data of Table I and Figs. 1–5 show a number of interesting features. Perhaps the most striking of these is the change in the character of the mass distribution on going from deuterons to helium ions as bombarding projectiles. Assuming that most of the fission comes from complete capture of the bombarding projectile, the former give actinium nuclei the latter thorium nuclei. At comparable initial excitation energies, e.g., 21.5-Mev deuterons vs 31-Mev He ions, where initial excitation energies are in the neighborhood of 30 Mev the deuteron-induced fission results in predominantly symmetric fission, whereas He ion-induced fission is distinctly asymmetric in character. This is in accord with the suggestion that asymmetric fission is dying out as a mode of fission in the element region below thorium.[7] A larger fraction of the total cross section goes into fission in the thorium nuclides (Table II) and this fission is predominantly asymmetric, compared with actinium nuclides. Somewhere in the region of nuclei between astatine and actinium asymmetric fission disappears altogether, because nuclides in the region from thallium to astatine apparently fission symmetrically.[7,8]

From the mass-yield curves observed for fission induced in Ra^{226} by protons and deuterons fission in actinium nuclei evidently gives rise to separate symmetric and asymmetric mass divisions. For these nuclei fission represents a smaller fraction of the total cross section, and it is evidently asymmetric fission which is suppressed, thereby allowing symmetric fission to stand out by itself. Symmetric fission could conceivably be present as a separate mode of mass division in the heaviest elements, but because of the greater preponderance of asymmetric fission in these elements, coupled with the narrower valley between the asymmetric wings,

[5] J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley and Sons, New York, 1952), pp. 352, 353.
[6] M. Shapiro, Phys. Rev. **90**, 171 (1953).

[7] A. W. Fairhall, R. C. Jensen, and E. F. Neuzil, reference 3, Vol. 15, p. 452, Paper 677.
[8] E. F. Neuzil, Ph. D. thesis, Department of Chemistry, University of Washington, 1959 (unpublished).

any such separate symmetric mode would be difficult to observe.

A comparison of the mass-yield curves for 21.5-Mev and 14.5-Mev deuteron-induced fission of Ra^{226} reveals that there is an enhanced yield of fragments from symmetric fission at the higher energy. Also, fission represents a somewhat larger fraction of the total cross section at the higher energy. This is in keeping with the pattern for symmetric fission observed in both the light element region below astatine and in the heaviest elements: the probability of symmetric fission increases with increasing excitation energy up to initial excitation energies at least as high as 35 Mev.[7,8] From studies of the energy and mass number dependence of symmetric fission of the several lead isotopes[8] it was observed that approximately 80% of the symmetric fission observed at any initial excitation energy up to about 35 Mev occurs before evaporation of any neutrons. Most of the remaining 20% of the symmetric fission occurs after the evaporation of the first neutron from the initial compound nucleus.

On this basis we attribute the central hump in the mass-yield curves of deuteron-induced fission of Ra^{226} to fission of Ac^{228}, and to a lesser extent to Ac^{227}. The asymmetric fission we attribute to fission of Ac^{226} and Ac^{225} nuclei excited to energies in the neighborhood of 10 Mev or less. At excitation energies near 10-Mev fission in these nuclei is expected to be asymmetric, in keeping with the behavior of the heaviest elements at this region of excitation energies.

In this interpretation we make use of separate symmetric and asymmetric modes of mass division with the distinction being made on the basis of the energy which the nucleus has when it undergoes fission. The experiment using 14.5-Mev deuterons to bombard Ra^{226} was an attempt to verify this procedure experimentally. At this particular bombarding energy, if the compound nucleus Ac^{228} loses a neutron then one gets Ac^{227} at about the same excitation energy as in the experiment where Ra^{226} is bombarded with 11-Mev protons. In principle a careful comparison of the fission cross sections and mass yield curves for 14.5-Mev deuterons vs 11-Mev protons would reveal how much fission occurred in Ac^{228} prior to neutron evaporation, and what the mass distribution of this prompt fission was. Unfortunately uncertainties in the values of the fission cross sections and in the total reaction cross sections in these experiments are too large to make this comparison possible. A comparison of the mass-yield curves in the two cases reveals that there is somewhat more symmetric fission relative to asymmetric fission in the deuteron experiment relative to the proton experiment, which is qualitative evidence that prompt fission is symmetric in character. However, it cannot establish how much, if any, asymmetric fission occurs at the higher energies.

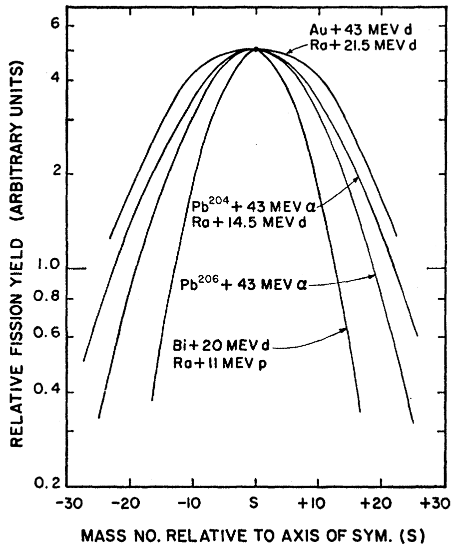

FIG. 6. Several mass distribution curves superimposed along their respective symmetry axes so as to display the variations in their widths.

It seems worthwhile to repeat this experiment. To do it properly would require protons of energy higher than 11 Mev in order to be above the energy region where the total reaction cross section is changing rapidly with energy.

One final observation which is worth pointing out concerns the width of the central peak, representing symmetric fission, for fission induced in Ra^{226} by deuterons. It is significantly wider than either the central peak of the mass distribution from fission induced in Ra^{226} by 11-Mev protons, or the single symmetric peak for fission induced in Bi^{209} by 22-Mev deuterons.[9] Recent studies of 43-Mev helium-ion induced fission of Au^{197}, Pb^{204}, and Pb^{206} also show differences in the widths of the symmetric mass-yield distributions observed for these target species.[8] Figure 6 shows a superposition of the symmetric fission portion of the mass distributions for these several cases. The variations in width of these curves is apparent. It is not possible to tell at the present time whether the variation in the widths of symmetric mass distributions is to be associated in some way with the excitation energies which the fissioning species have, or whether it represents intrinsic properties of the nuclear species which undergo fission. Further work is in progress to try to learn more about this question.

ACKNOWLEDGMENTS

We wish to thank Ted J. Morgan and the crew of the University of Washington cyclotron for their assistance in making bombardments.

[9] A. W. Fairhall, Phys. Rev. **102**, 1335 (1956).