line becomes apparent.[24] Some of the discrepancy may also be due to errors in the experimental results for $E_K{}^*$ or ν values. The uncertainty in $E_K{}^*$ is estimated to be about ± 2 MeV; however, the uncertainty in absolute values of ν_T (as distinguished from point-to-point uncertainties) may be as high as ± 1 neutron, corresponding to about 8 MeV in E_T.

In discussing total energy balance thus far, the possible existence of two fission components has not been considered. In the two-component picture, we would have the same total reaction energy E_T as shown in Fig. 13 for each component, although the kinetic and excitation energies would be divided somewhat differently. We have indicated in Fig. 13 (with light solid lines) the total kinetic and total excitation energies for the two components as obtained from decomposition, as discussed above. The appropriate portions of the discussion of Fig. 11 also apply to these curves.

[24] This point has been discussed in Ref. 21; references to other mass formulas are contained in that paper. There is no particular reason to prefer the Wing-Fong formula; we use it here for consistency with earlier work in low-excitation fission. See Ref. 21; also J. H. Neiler, F. J. Walter, and H. W. Schmitt, Phys. Rev. **149**, 894 (1966).

ACKNOWLEDGMENTS

The authors acknowledge with appreciation the assistance of Miss Frances Pleasonton and R. W. Lide in operation of the experiment, the assistance of Mrs. B. H. Hannon and C. W. Nestor in processing the data, and the cooperation of G. F. Wells and the Oak Ridge tendem staff in operation of the accelerator. Helpful discussions with J. R. Nix, F. Plasil, and R. L. Ferguson are gratefully acknowledged. One of us (E. K.) gratefully acknowledges a fellowship from the German Bundesministerium für wissenschaftliche Forschung. He also appreciatively acknowledges the hospitality of the Oak Ridge National Laboratory during his one-year visit there.

PHYSICAL REVIEW VOLUME 172, NUMBER 4 20 AUGUST 1968

Asymmetry, Anisotropy, and Excitation Function for the Proton-Induced Fission of ^{226}Ra†

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(Received 11 March 1968)

The anisotropy of the fragment angular distribution, the asymmetry of the fragment mass distribution, and the relative cross section for proton-induced radium fission have been measured as functions of incident proton energy in the range $6.0 \leq E_p \leq 13.0$ MeV. The anisotropy and excitation function have also been studied as functions of E_p for the symmetric and asymmetric components of the mass distribution. The angular-distribution data for the two components of the mass distribution are similar. There is no conclusive evidence in these data to indicate whether one or two saddle-point states are involved in Ra+p fission.

I. INTRODUCTION

A DETAILED study of the kinetics and of the neutron-emission data for proton-induced fission of ^{226}Ra, which is presented in the preceding paper,[1] indicates that the notion of two components may be used to describe radium fission. The distribution centered about symmetric mass division appears to be describable in the general form of the liquid-drop theory[2]; asymmetric fission shows features similar to those appearing in the low-excitation fission of heavy nuclei and seems to be strongly influenced by the nuclear properties of the fragments.

It has remained unclear, however, whether these two possible components are associated with two separate saddle-point states or whether they develop at a later stage of the fission process, e.g., in the descent from the saddle-point to scission. In an attempt to obtain some insight into this problem, we have carried out detailed studies of the anisotropy, the asymmetry, and the relative cross section for radium fission as functions of incident-proton energy in the range $6.0 \leq E_p \leq 13.0$ MeV. Previous studies related to this work include those of Gindler, Bate, and Huizenga,[3] in which the fragment angular distributions in charged-particle fission of radium were studied. Also, some information concerning the anisotropy and asymmetry was obtained by Schmitt, Dabbs, and Miller.[4] The

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† Research sponsored by the U. S. Atomic Energy Commission under contract with the Union Carbide Corp.

[1] E. Konecny and H. W. Schmitt, preceding paper, Phys. Rev. **172**, 1213 (1968).
[2] J. R. Nix and W. J. Swiatecki, Nucl. Phys. **71**, 1 (1965).
[3] J. E. Gindler, G. L. Bate, and J. R. Huizenga, Phys. Rev. **136**, B1333 (1964).
[4] H. W. Schmitt, J. W. T. Dabbs, and P. D. Miller, in *Proceedings of the Symposium on the Physics and Chemistry of Fission Salzburg, 1965* (International Atomic Energy Agency, Vienna, 1965), Vol. I, p. 517.

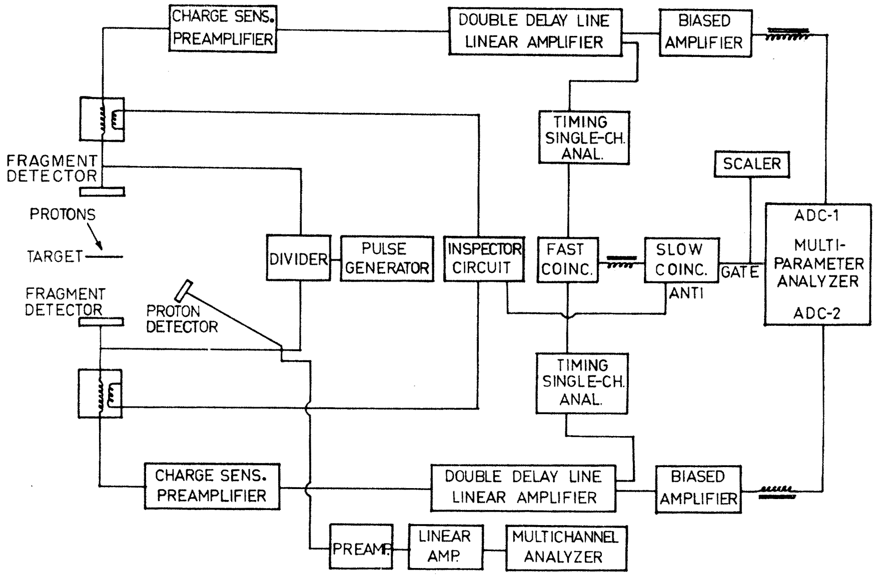

FIG. 1. Schematic diagram of the experimental arrangement.

asymmetry has been studied by means of radiochemical yield measurements by Jensen and Fairhall,[5] by Wolke,[6] and recently by Bowles and Beckett.[7] In the present work we have studied the anisotropy and relative fission cross section for all fragments together and also for the symmetric and asymmetric distributions separately.

II. METHOD AND APPARATUS

Protons of energy E_p in the range 6.0 to 13.0 MeV were obtained at the Oak Ridge tandem Van de Graaff accelerator. The proton beam impinged on a thin ($\sim$40 μg/cm^2) target of ^{226}Ra bromide, which was vacuum-evaporated onto a 20-μg/cm^2 carbon backing. Pulse heights from coincident fission fragments incident on two silicon surface-barrier detectors were recorded. A third surface-barrier detector with a relatively deep ($\sim$2 mm) depletion layer was employed to monitor the elastically scattered protons. This detector was mounted at a fixed angle of 30° with respect to the proton-beam direction and at a distance of 100 cm from the target. Collimating apertures were used to reduce background from scattered protons.

The fission-fragment detectors were mounted on-axis with the target and were located at a distance of 7.5 cm from the target. The angle of the detector-target-detector axis could be varied in the range 30° to 150° with respect to the beam direction.

The experimental arrangement is shown schematically in Fig. 1. Double delay-line linear amplifiers were used, and a fast coincidence was established at the crossover point of the bipolar pulses. Reduction of pileup pulses (α-on-fission and proton-on-fission) was accomplished by means of the time-pickoff and inspector circuits, together with the anticoincidence requirement in the slow coincidence circuit. Correlated fragment pulse heights were recorded event by event in 256×256 channels on punched paper tape. The total number of accepted fission events (gate signals at the analyzer) was counted independently by a digital scaler as shown.

An independent multichannel analyzer was used to record the energy spectrum of protons scattered from the target; the peak corresponding to elastically scattered protons from radium bromide was selected and used to monitor the relative number of incident protons.

III. DATA AND ANALYSIS

Two different sets of measurements were carried out. In one set, consisting of many short ($\sim$10 min) runs, a single detector was used to detect and count the total number of fragments as a function of angle of emission and as a function of proton energy. For this purpose the double-coincidence requirement and inspector functions were omitted, inasmuch as the occurrence of α-on-fission or proton-on-fission pileup was not important, and pulses smaller than those associated with fission fragments could be successfully discriminated against.

The second set of measurements, consisting of longer two-parameter runs with the arrangement as shown in Fig. 1, was carried out at selected proton energies and at selected angles in order to study the relative behavior of the symmetric and asymmetric components of radium fission. About 20 000 events were included in each such two-parameter measurement.

Data analysis for the two-parameter experiments proceeded according to the method outlined in a pre-

[5] R. C. Jensen and A. W. Fairhall, Phys. Rev. **109**, 942 (1958); **118**, 771 (1960).
[6] R. L. Wolke, Phys. Rev. **120**, 543 (1960).
[7] B. J. Bowles and N. Beckett, Phys. Rev. **147**, 852 (1966).

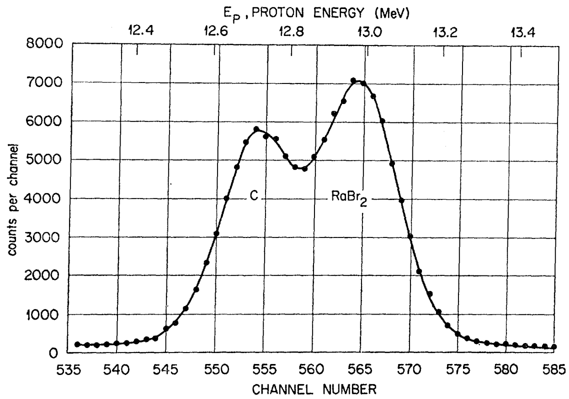

FIG. 2. Energy spectrum of elastically scattered protons. $E_p=13.0$ MeV, $\theta=30°$. The two groups result from scattering from radium bromide and from the carbon backing.

vious publication.[8] Since the object of the present experiments was only to determine the relative numbers of events occurring in the symmetric and asymmetric distributions, no neutron-emission effects were taken into account, and the analysis was carried only far enough to obtain the distribution $N(\mu_1)$ of provisional masses μ_1. It may be recalled that $\mu_1=AE_2/(E_1+E_2)$, where E_1 and E_2 are the measured single-fragment kinetic energies corrected for center-of-mass motion and A is the mass of the fissioning nucleus; the mass-dependent energy calibration of fragment pulse heights[9] is included in the analysis.

Since detailed fragment spectroscopy was not essential in these measurements, the fission detectors were not required to have extremely good resolution throughout. Some slight deterioration of detector resolution was observed during the course of the measurements, principally because of the relatively large dose of fission fragments. Appropriate checks were made throughout the runs to monitor the effect and to assure sufficient reliability (for present purposes) of the mass-distribution data.

Inasmuch as the mass distributions $N(\mu)$ were observed to be symmetric within experimental uncertainties, it was possible to improve statistical errors (for subsequent fitting) by symmetrization, i.e., addition of counts occurring at masses μ and $A-\mu$. One-half of the mass distribution could then be fitted with two Gaussian distributions or, equivalently, the full mass distribution with three Gaussian distributions. The ratio $W_{\text{sym}}/W_{\text{asym}}$ is then the ratio of total area of the outer distributions to that of the inner distribution.

More correctly, from the point of view of the two-component picture, only the central peak should be fitted with a Gaussian distribution; the ratio of this area to the remaining area would then give $W_{\text{sym}}/W_{\text{asym}}$. Such calculations were carried out for distributions obtained for the low, intermediate, and high incident proton energies of the experiment, with results that agreed within 1 or 2% with the results of the three-Gaussian fitting procedure. For consistency, and to remove uncertainties (a few percent) arising from the arbitrary choice of cutoff mass in the single-Gaussian fit, the three-Gaussian fit was used throughout the analysis.

The product of proton intensity and the number of available radium atoms was measured by means of the third solid-state detector, in which protons elastically scattered from the radium-bromide target at an angle of 30° with respect to the beam direction were monitored.

The detector resolution was sufficiently good to permit the separation of the proton groups elastically scattered from carbon and from radium bromide,

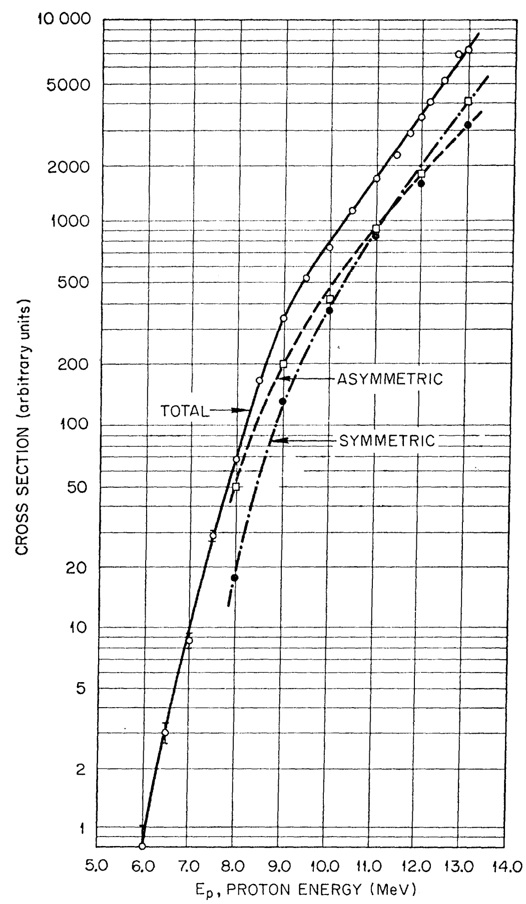

FIG. 3. Excitation function for proton-induced fission of ^{226}Ra. The solid line shows the total cross section; the symmetric and asymmetric components are shown by the dot-dashed and dashed curves, respectively. Error bars referring to the statistical error are shown where the error exceeds the size of the data points. An estimate of the absolute cross section gives 8.5 ± 3.5 mb for the total fission cross section at $E_p=11.0$ MeV.

[8] H. W. Schmitt, J. H. Neiler, and F. J. Walter, Phys. Rev. **141**, 1146 (1966).

[9] H. W. Schmitt, W. E. Kiker, and C. W. Williams, Phys. Rev. **137**, B837 (1965); H. W. Schmitt, W. M. Gibson, J. H. Neiler, F. J. Walter, and T. D. Thomas, in *Proceedings of the Symposium on the Physics and Chemistry of Fission, Salzburg, 1965* (International Atomic Energy Agency, Vienna, 1965), Vol. I, p. 531.

although it was not possible to separate the groups scattered from Ra and from Br. Figure 2 shows a typical energy spectrum of elastically scattered protons for $E_p = 13$ MeV. The separation of the two components was accomplished by fitting the experimental curves with two Gaussian distributions.

The Rutherford formula was used for the calculation of the proton scattering cross section, since the distances of closest approach in elastic scattering of protons by heavy atoms are larger than the nuclear radii for the energy range of the present experiment. The differential fission cross section at an angle θ with respect to the proton beam is given by

$$(d\sigma/d\Omega)_{f,\theta} = [N_f(\theta)/2N_p](d\Omega_p/d\Omega_f)(d\sigma/d\Omega)_p, \quad (1)$$

where $N_f(\theta)$ and N_p are the fission-particle rate and the proton rate, respectively; $(d\sigma/d\Omega)_p$ is the Rutherford scattering cross section for protons on $RaBr_2$ at 30° to the beam.

The total fission cross section may be obtained by integrating over the full solid angle. Since the anisotropy is small (as will be discussed in Sec. IV), the integral is not much different from $4\pi(d\sigma/d\Omega)_{f,\theta=90°}$; it is larger at most by a factor of ~ 1.04 in the present experiments. Relative cross sections will be discussed in some detail; absolute cross sections, however, were determined only approximately.

IV. RESULTS

A. Excitation Function and Asymmetry

The excitation function for proton-induced fission of radium is given in Fig. 3. Curves are shown for the total fission cross section, measured as indicated in Sec. III, and for the symmetric and asymmetric components, determined with the aid of two-parameter experiments carried out at selected energies throughout the full range. An estimate of the absolute cross section, determined for $E_p = 10.5$ MeV, gives $\sigma_f = 5.5 \pm 2.5$ mb. This value is essentially in agreement with the value 4.6 ± 2.2 mb given by Gindler, Bate, and Huizenga[3] for protons of the same energy. Our estimate gives $\sigma_f = 8.5 \pm 3.5$ mb for $E_p = 11.0$ MeV, compared with 2 ± 1 mb given by Jensen and Fairhall[5] from early measurements at this energy.

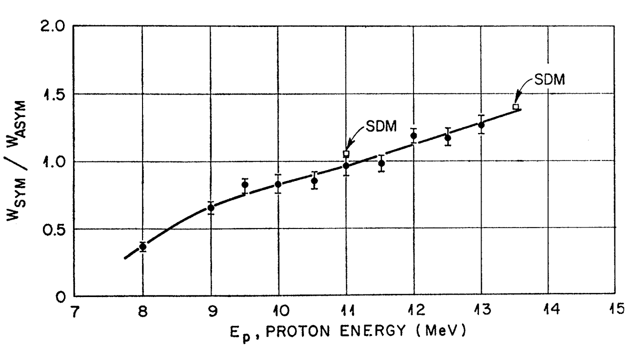

FIG. 4. Ratio of the yield of the symmetric component W_{sym} to that of the asymmetric component W_{asym} as a function of proton energy. Open squares labeled SDM are from Ref. 4.

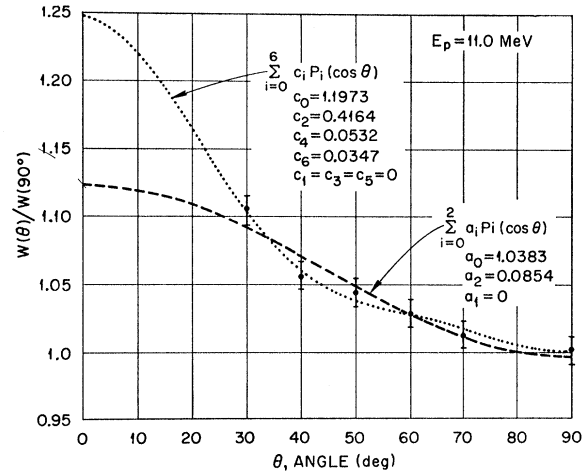

FIG. 5. Fission-fragment angular distribution for $E_p = 11.0$ MeV. Dotted and dashed curves show least-squares fits of the data to sixth- and second-order Legendre polynomials, respectively. See text.

The observed total fission excitation function shows a steep increase in relative fission cross section with increasing proton energy, as expected on the basis of Coulomb barrier penetration. The excitation function is found to be less steep than that reported by Jensen and Fairhall[5] but is in fairly good agreement with the result of Wolke.[6] We find the ratio of the cross section at $E_p = 11.0$ MeV to that at $E_p = 9.0$ MeV to be 5.2, compared with the value 60 of Ref. 5, and the ratio of the cross section at $E_p = 13.6$ MeV (extrapolated) to that at $E_p = 11.0$ MeV to be ~ 6.4, compared with the value 7.1 of Ref. 6.

The mass asymmetry as a function of proton energy is shown in Fig. 4. The present data are in good agreement with the corresponding data as calculated from the yield curves of Ref. 4.

At higher proton energies Bowles and Beckett[7] found a stepwise behavior of the asymmetry as a function of proton energy superimposed on the generally stronger perference for symmetric fission with increasing energy. Whenever the compound-nucleus excitation energy is sufficient to emit one or more neutrons prior to fission, and when such neutron emission occurs, the compound-nucleus excitation energy is decreased by an amount equal to the binding plus kinetic energy of the neutron, thus giving rise to an enhancement of asymmetric fission. Although we have drawn a smooth curve through the data points in Fig. 4, it is possible that a small effect of this type occurs at about 10 MeV. We have estimated, by the method of Huizenga and Vandenbosch,[10] the probability of second-chance fission

[10] J. R. Huizenga and R. Vandenbosch, in *Nuclear Reactions*, edited by P. M. Endt and P. B. Smith (North-Holland Publishing Co., Amsterdam, 1962), Vol. II.

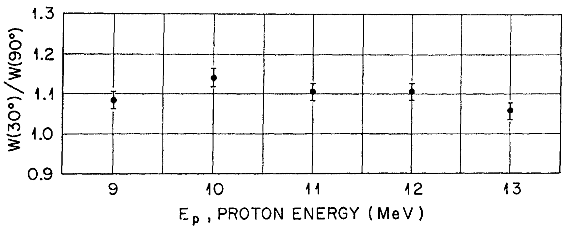

FIG. 6. Fission-fragment anisotropy $W(30°)/W(90°)$ as a function of incident-proton energy.

relative to that of primary fission and have found it to be only a few percent in the present energy range.

B. Fission-Fragment Angular Distributions

The usual theory for the description of fission-fragment angular distributions is based on considerations of the collective rotations and oscillations of the compound nucleus.[11–13] The angular distribution is determined by the particular excited state at the saddle point through which fission occurs; these states are characterized by the total angular momentum I and its projection K on the nuclear symmetry axis. Much experimental evidence indicates that these quantities remain good constants of motion in the descent from the saddle point to scission. According to the theory, the distribution in K is a Gaussian distribution centered at $K=0$ and characterized by the quantity K_0^2, the average value of K^2, given by

$$K_0^2 = tJ_{\text{eff}}/\hbar^2, \quad (2)$$

where t is the nuclear temperature, $J_{\text{eff}} = J_{\parallel}J_{\perp}/(J_{\parallel}+J_{\perp})$ is the effective moment of inertia, with $J_{\perp}$ and $J_{\parallel}$ the moments of inertia with respect to axes perpendicular and parallel, respectively, to the axis of deformation. The anisotropy is given to good approximation by

$$W(0°)/W(90°) = 1 + \bar{I}^2/4K_0^2, \quad (3)$$

where $\bar{I}^2$ is the average of the square of the spin. For the present qualitative discussion, we note that for a given proton energy (and thus constant $\bar{I}^2$) the anisotropy depends on the nuclear shape (through the moments of inertia) and on the transition-state energy (through the nuclear temperature). For small values of K_0^2 the theory predicts anisotropic fragment distributions which peak toward 0° and 180°; for larger values of K_0^2 the theory predicts more nearly isotropic distributions.

Let us expand the angular distribution in the usual way as a sum of Legendre polynomials:

$$W(\theta)/W(90°) = \sum_i \alpha_i P_i(\cos\theta). \quad (4)$$

The experimental determination of the particular components α_i of the fission-fragment angular distribution suffers somewhat from the fact that measurements could not be extended to angles below 30° in the present apparatus. Illustrative of this point is the observation that the extrapolated value of $W(0°)/W(90°)$ depends on the cutoff applied to the Legendre expansion, and depends most sensitively on the data point at $\theta=30°$. The angular-distribution data for 11.0-MeV proton-induced fission are shown in Fig. 5 as an example. These data have been fitted by least-squares fits to a sum of Legendre polynomials up to second and sixth orders, as shown by the smooth curves in the figure. Although the sixth-order polynomial fit is somewhat better than the second-order fit, the latter may not be excluded on the basis of one such set of data. It is found, however, that for all proton energies investigated, the point at $\theta=30°$ is displaced upward from the second-order fit, indicating that higher-order terms should probably be included. This finding is consistent with previous studies for other fissioning nuclei. We note also that both of the fits shown in Fig. 5 result in extrapolated anisotropies somewhat higher than the value of $W(174°)/W(90°)=1.07\pm0.03$ for 10.5-MeV protons reported by Gindler *et al.*[3]

In Fig. 6 the measured quantity $W(30°)/W(90°)$ is plotted as a function of incident-proton energy. The magnitude of the anisotropy so defined is seen not to change appreciably with proton energy within the energy range investigated.

It is of interest to determine whether the angular distributions are different for the symmetric and asymmetric components of the mass distribution. Figure 7 shows the quantity $W_{\text{sym}}/W_{\text{asym}}$ as a function of the angle θ between the direction of the fission fragments and the proton beam, with the proton energy as a parameter. Within the experimental error, no correlation between $W_{\text{sym}}/W_{\text{asym}}$ and θ is apparent for 9 MeV$\leq E_p \leq$13 MeV. We have, however, also incorporated the corresponding data, Ref. 4, for 13.5-MeV protons. Those data, reported in terms of the ratios of yields in the symmetric and asymmetric peaks, have been converted to the ratios of areas under the respective peaks, for consistency with the other curves. The small correlation of asymmetry with anisotropy which seems to appear in this curve was attributed to a small contribution of second-chance fission, for which the anisotropy of the asymmetric component is enhanced relative to that of the symmetric component.

V. DISCUSSION

On the basis of the experimental material presented above, an unambiguous decision as to whether two

[11] A. Bohr, in *Proceedings of the United Nations International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1955), Vol. 2, p. 151.

[12] I. Halpern and V. M. Strutinski, in *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva 1958* (United Nations, Geneva, 1958), Vol. 15, p. 408.

[13] J. Griffin, Phys. Rev. **116**, 107 (1959); **127**, 1248 (1962).

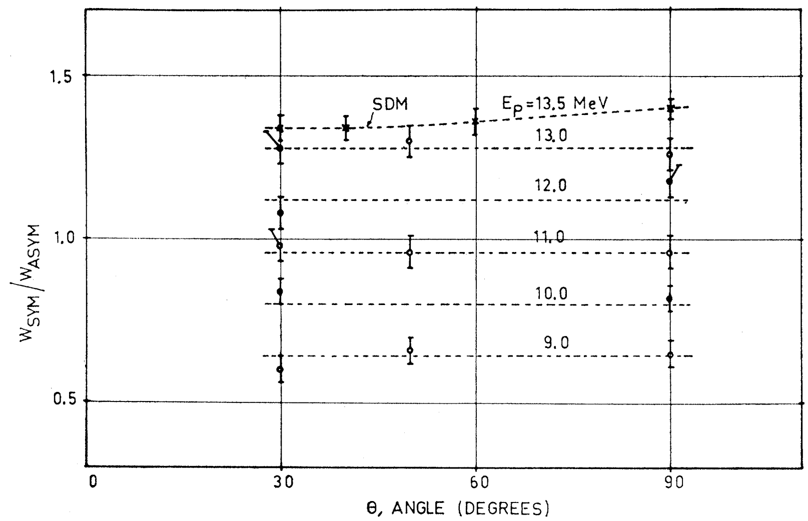

FIG. 7. Ratio of symmetric to asymmetric yields W_{sym}/W_{asym} as a function of the direction of fragment emission with respect to the proton beam, for various proton energies. The corresponding data of Ref. 4, $E_p = 13.5$ MeV, are included.

distinct saddle points are associated with the two components of the mass distribution does not appear to be possible. The decomposed excitation functions shown in Fig. 3 seem to indicate different behavior near threshold for asymmetric and symmetric fission. This difference, however, cannot be taken as a strong indication that two different saddle energies occur, since the observed increase in relative probability of asymmetric fission at low excitation may also be the result of a preference for asymmetric fission at a later stage of the fission process.

A more reliable conclusion might be drawn from the angular-distribution data. On the basis of Eq. (3), one would expect that, if there are two saddle-point states involved, and if their shapes or energies are different, then the angular distribution would be different for the two components. Thus the ratio W_{sym}/W_{asym} would vary with angle, contrary to the results shown in Fig. 7.

Therefore, if there are two saddle-point states, their characteristics must be quite similar. We are able, then, to conclude only that either (a) a single saddle point is involved, or (b) two saddle points with very similar properties are involved in Ra+p fission. This leaves the situation essentially unchanged in that, so far as we are aware, no experimental evidence has been found which conclusively indicates the existence of two different saddle-point states for symmetric and asymmetric fission, respectively.

ACKNOWLEDGMENTS

The authors acknowledge with appreciation the assistance of Miss Frances Pleasonton in various aspects of this work, the assistance of Mrs. B. H. Hannon and C. W. Nestor in processing the data, and the cooperation of G. F. Wells and the Oak Ridge tandem staff in operation of the accelerator. One of us (E. K.) gratefully acknowledges a fellowship from the German Bundesministerium für wissenschaftliche Forschung. He also appreciatively acknowledges the hospitality of the Oak Ridge National Laboratory during his one-year visit there.