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Angular Distribution of Fragments from the Fission of Bismuth by 450-Mev Protons*†

ROBERT L. WOLKE‡ AND JOEL R. GUTMANN§
Enrico Fermi Institute for Nuclear Studies, University of Chicago, Chicago, Illinois

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The angular distributions of three fragments from the 450-Mev proton-induced fission of Bi have been measured by a recoil-catching technique involving radiochemical separation of the fission products. The forward velocity component of the fissioning nucleus was determined relative to the velocity of each fragment. If it is assumed that the motion of the fissioning nucleus is directed along the proton axis, the angular distributions in the system of the fissioning nucleus are found to be of the form $a+b\cos^2\theta'$, with values of b/a of 0.10±0.01, 0.115±0.015, and 0.09±0.01 for $Ga^{72,73}$, $Sr^{91,92}$, and $Cd^{115,117}$, respectively. The possibility that the motion of the fissioning nucleus is directed at an angle to the proton axis is discussed.

I. INTRODUCTION

THE angular distributions of fragments from the fission of several heavy elements have been measured previously by a variety of techniques. Although the fission of U^{235} by thermal neutrons has been shown to be isotropic,[1] the fragments from fission induced by energetic particles or electromagnetic radiation tend to be emitted preferentially (in the center-of-mass system) either parallel to, or perpendicular to, the direction of the exciting radiation. A recoil-catching method has shown[2,3] that fragments from the fission of thorium and uranium by bremsstrahlung of maximum energy from 8 to 16 Mev favor perpendicular emission, with angular distributions of the form $a+b\sin^2\theta$, where θ is the angle between the fission fragment and the direction of the incident radiation. The anisotropy parameter, b/a, was found to decrease as the maximum bremsstrahlung energy was raised from 8 to 16 Mev, and to increase with increasing mass ratio of the fragments. Recoil-catching techniques have also been employed to measure the angular distributions of specific fragments from the 22-Mev proton-induced fission of thorium and several uranium isotopes.[4,5] The center-of-mass distributions were found to be of the form $a+b\cos^2\theta$, indicating a preference for parallel emission. The anisotropy was found to be smallest for the symmetric fission products, increasing with the mass asymmetry of the observed fragment. The fast neutron-induced fission of several heavy nuclides has been investigated in an ionization chamber by Brolley and co-workers,[1,6] who found that when U^{235} is exposed to neutrons in the energy range from 0 to 20 Mev, the fission fragments follow an $a+b\cos^2\theta+c\cos^4\theta$ distribution function, with the $\cos^4\theta$ term dominating. Similar results were obtained when the fission of Np^{237} was induced by 14-Mev neutrons.

The present set of experiments was undertaken in order to determine whether the parallel anisotropies observed in the low-energy particle-induced fission of heavy nuclides exist also for a lighter target nucleus at higher bombarding energies. Since the present work was begun, however, it has become apparent[7–9] that the type of anisotropy exhibited by fast-particle-induced fission depends both upon the energy of the exciting particle and upon the nucleus being bombarded. Recent work by Henkel and Brolley,[7] for example, has shown that although Th^{232} exhibits a parallel anisotropy of neutron-induced fission over most of the neutron energy range from threshold to 10 Mev, perpendicular fission is preferred in the region of $E_n\approx 1.6$ Mev. Lozhkin and co-workers[8] have shown that while the fission of U^{238} by neutrons of energies up to 20 Mev occurs preferentially parallel to the beam,[7] the gross fission fragments from the 660-Mev proton-induced fission of natural uranium are preferentially emitted perpendicular to the beam. The distribution at 660 Mev was found to be given by the function $a+b\sin^4\varphi$, where φ is the angle between the incident proton direction and the projection of the fragment track in the plane of a loaded nuclear emulsion oriented edgewise to the beam. Recent recoil studies[9] in this laboratory, while confirming the preference for parallel emission of fragments from the 450-Mev proton-induced fission of bismuth reported herein, have shown that fission fragments from tantalum targets exposed to 450-Mev protons are emitted either iso-

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† Preliminary reports of the present work were presented before the 1955 New York and 1956 Washington meetings of the American Physical Society: R. L. Wolke, Phys. Rev. **98**, 1199 (1955) and Bull. Am. Phys. Soc. Ser. II, **1**, 165 (1956).

‡ Present address: Department of Chemistry, University of Florida, Gainesville, Florida.

§ On leave of absence from the Israel Atomic Energy Commission. Present address: Weizmann Institute of Science, Rehovoth, Israel.

[1] J. E. Brolley, Jr., and W. C. Dickinson, Phys. Rev. **94**, 640 (1954).

[2] Winhold, Demos, and Halpern, Phys. Rev. **87**, 1139 (1952).

[3] E. J. Winhold and I. Halpern, Phys. Rev. **103**, 990 (1956).

[4] Cohen, Jones, McCormick, and Ferrell, Phys. Rev. **94**, 625 (1954).

[5] Cohen, Ferrell-Bryan, Coombe, and Hullings, Phys. Rev. **98**, 685 (1955).

[6] Brolley, Dickinson, and Henkel, Phys. Rev. **99**, 159 (1955).

[7] R. L. Henkel and J. E. Brolley, Jr., Phys. Rev. **103**, 1292 (1956).

[8] Lozhkin, Perfilov, and Shamov, J. Exptl. Theoret. Phys. U.S.S.R. **29**, 292 (1955) [translation: Soviet Phys. JETP **2**, 116 (1956)].

[9] N. Porile and N. Sugarman, Bull. Am. Phys. Soc. Ser. II, **1**, 328 (1956), and (to be published).

tropically or preferentially perpendicular to the beam direction.

The present paper reports the determination of the angular distributions in the center-of-mass system of three specific fragments from the fission of Bi^{209} by 450-Mev protons. Fragments escaping from a spherical bismuth target were caught with efficient geometry on an aluminum cone which was subsequently cut into pieces corresponding to various recoil angles, and subjected to radiochemical separations. From activity measurements of the separated fission products, the relative numbers of fragments emitted in various directions were determined and the angular distributions were calculated.

II. EXPERIMENTAL PROCEDURE

The target assembly (Fig. 1) consisted essentially of a spherical source of fission fragments supported on the axis of a conical recoil-catcher. The source was a $\frac{3}{32}$-inch brass sphere onto which was electroplated about 15 mg/cm^2 of bismuth, and which was supported on a holder by a 12-mil copper wire soldered into it. The thickness of the bismuth coating was greater than the range of any of the fission fragments observed. The recoil-catcher was a partial conical surface, 3 cm in altitude and 6 cm in base diameter, of very pure 1-mil aluminum foil.[10] The cone was supported at its base by a circular arc of brass spring wire, which clipped onto the chromium-plated brass holder. The entire assembly was mounted on the end of a 4-inch-diameter probe and inserted into the circulating proton beam of the synchrocyclotron, with the spherical target in the median plane and the holder oriented so that the beam traveled along the axis of the cone. A segment of the cone was missing, in order to provide an almost unhindered spiral proton path from the center of the cyclotron to the Bi sphere.

Separate sets of runs were made to measure the forward and backward angular distributions. Figure 1 shows the assembly oriented to catch the fragments emitted at laboratory angles of from 0° to 90° to the beam. For the backward runs, the probe was rotated 180°, so that the apex of the cone pointed "upstream," and fragments emitted at laboratory angles of 90° to 180° were caught on the cone.

After each bombardment, the catcher-cone bearing the fission-fragment recoils was removed and cut into circular zones corresponding to 30°-wide intervals of recoil angle with respect to the direction of the proton beam. The pieces of Al foil were then dissolved and subjected to radiochemical fission-product separations.[11] The samples were counted by end-window methane-flow proportional counters. All measured activities were corrected for chemical yield and self-absorption.

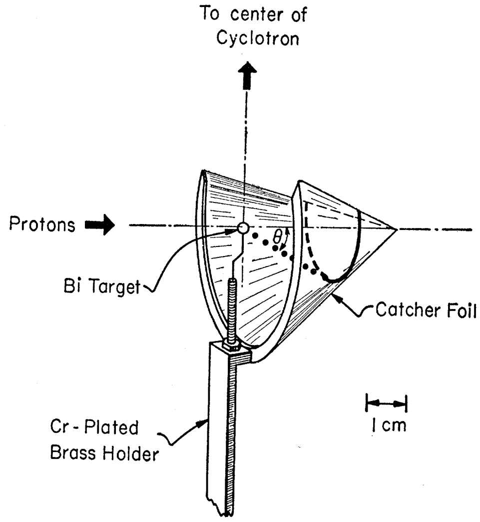

FIG. 1. Target and recoil catcher.

The foils were cut to correspond to a given recoil angle θ with an estimated precision of one degree near $\theta=0°$ and 180°, and 0.3 degree near $\theta=90°$. This precision is well within the resolution of the apparatus, which was limited to about 3° by the finite size of the spherical source. The errors from counting statistics were generally less than 1%. Chemical yields were determined to a precision of about 1% by weighing the mounted precipitates. Self-absorption curves were determined in a separate series of experiments, using the same precipitates and counting geometries used in the runs. These permitted the determination of self-absorption correction factors which were accurate to about 5%. Since the pertinent data consisted of activity *ratios*, and since the samples generally differed only slightly in weight, the errors due to self-absorption effects were less than 1%.

There were two sources of extraneous fission-product activity in the catcher foil. First, impurities in the brass holder, which were activated by scattered protons, were found to escape from the brass surfaces and be caught on the Al cone. This difficulty was overcome by electroplating all exposed brass surfaces with chromium. Secondly, impurities in the Al foil itself, which intercepted a significant amount of the circulating beam due to vertical and radial oscillations, were activated. The amounts of such extraneous activity varied for different fission products, from less than 1% of the total activity on the foil for Sr to about 15% for Cd. A correction for this spurious activity was made by measuring the ratio of the impurity activation in the Al foil to the induced Na^{24} activity during a series of "blank runs" in which the brass bead was plated with chromium instead of bismuth. This ratio remained constant to less than 1% throughout the course of the work. The Na^{24}

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[10] 99.99% purity; generously supplied by the Aluminum Company of America.

[11] The chemical procedures employed were modifications of those given by W. W. Meinke, University of California Radiation Laboratory Report UCRL-432, 1949 (unpublished).

activity induced in each cut of the catcher foil was measured in each run to permit the calculation of the amount of "blank" activity to be subtracted.

Several possible sources of recoil anisotropy which might be inherent in the experimental design were investigated. Since the fission fragments emerge from the bismuth with a wide spectrum of charges and velocities, one might expect the difference in curvature of their paths by the magnetic field of the cyclotron to cause a serious distortion of the angular distributions. However, since the positive charge on a heavy ion of nuclear charge Z traversing matter is proportional to $Z^{\frac{1}{3}}$ times its velocity,[12] and since its radius of curvature is proportional to its velocity divided by its charge, the radius of curvature (projected on a plane perpendicular to the magnetic field) is constant for all recoils of a given atomic number, regardless of their velocities and ionic charges. The smallest radius of curvature of any fragment isolated in the present work (Ga^{72} traveling perpendicular to the magnetic field) was calculated to be 29 cm, a curvature which would produce a negligible displacement on the catcher in the 3 cm of ion flight between the Bi sphere and the cone.

The radial attenuation of the outward-spiralling protons by the target sphere might also be expected to introduce a serious anisotropy, since different parts of the sphere produce the recoils observed at different angles, and will receive different beam intensities depending on their distances from the center of the cyclotron. A mathematical analysis of the spherical recoil source yielded an expression for the "active volume," V_θ, of the sphere from which the recoils observed at an angle θ to the beam originate. The effect of a radially decreasing beam intensity on the observed angular distributions from such a source could then be calculated. The results showed that the same fraction of V_θ was subtended by any constant-beam section through the sphere, independent of θ. No perturbation of the angular distributions, therefore, is introduced by a radially varying beam intensity.

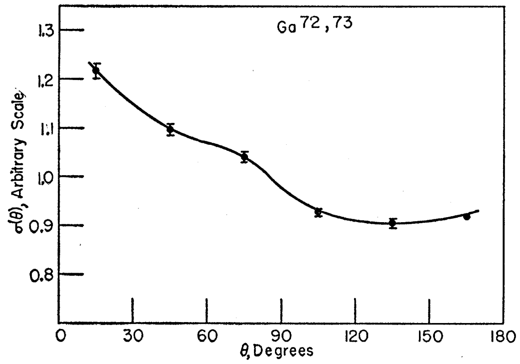

FIG. 2. Angular distribution of gallium fragments in the laboratory system.

III. TREATMENT OF DATA

The forward and backward angular distributions, which were measured in separate sets of runs, were normalized to the same number of fissions by first measuring the ratio of the total number of forward recoils to the total number of backward recoils in a set of experiments with stacked foils, and then calculating the corresponding ratio for a spherical geometry. For a *thick foil target* and isotropic emission of fragments in the system of the moving fissioning nucleus, it has been shown[13] that this ratio is given by

$$(\mathrm{ReF/ReB})_{\mathrm{foil}}=[(1+\eta)/(1-\eta)]^2, \qquad (1)$$

where ReF and ReB are the numbers of recoils emitted in the forward and backward hemispheres, respectively, and η is a measure of the forward center-of-mass motion. The quantity η is defined as the component of the fissioning-nucleus velocity along the proton axis divided by the velocity of the fission fragment in the system of the moving fissioning nucleus (hereafter referred to as the center-of-mass system).[14] For a *spherical target*, it can be shown that

$$(\mathrm{ReF/ReB})_{\mathrm{sphere}}=(1+\tfrac{3}{2}\eta+\tfrac{1}{3}\eta^2)/(1-\tfrac{3}{2}\eta+\tfrac{1}{3}\eta^2), \qquad (2)$$

where higher order terms in η have been neglected, since $\eta \ll 1$. The forward and backward runs can then be normalized by calculating η from the measured value of $(\mathrm{ReF/ReB})_{\mathrm{foil}}$ and using this value of η to obtain $(\mathrm{ReF/ReB})_{\mathrm{sphere}}$.

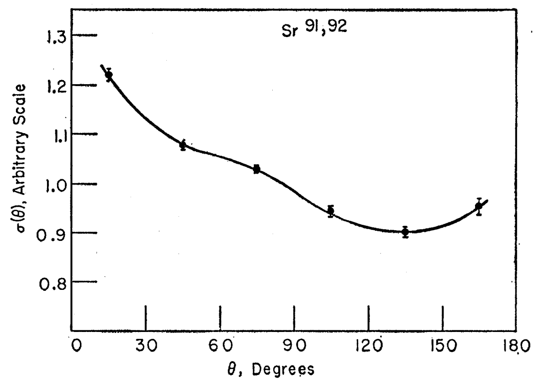

FIG. 3. Angular distribution of strontium fragments in the laboratory system.

Equations (1) and (2) assume an isotropic angular distribution in the center-of-mass system. Although they are only slightly different for an anisotropic center-of-mass distribution (as long as it is symmetric about 90°), the equations above should be modified to

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[12] N. Bohr, Phys. Rev. **59**, 270 (1941).

[13] Sugarman, Campos, and Wielgoz, Phys. Rev. **101**, 388 (1956).

[14] The system of the moving fissioning nucleus is not a true center-of-mass system, inasmuch as the incident proton is not included. We shall, however, use the designation "center of mass" in referring to the system of the moving fissioning nucleus. Similarly, the motion of the fissioning nucleus will be referred to as "center-of-mass motion."

account for center-of-mass anisotropy. The modified equations depend upon the functional form chosen to represent the distribution in the center-of-mass system. For a center-of-mass distribution of the form $a+b\cos^2\theta'$, where θ' is the angle between the proton axis and the direction of the fragment in the center-of-mass system, Eqs. (1) and (2) become

$$\left(\frac{\mathrm{ReF}}{\mathrm{ReB}}\right)_{\mathrm{foil}}=\frac{(1+\eta)^2+(b/a)(\frac{1}{2}+\frac{2}{3}\eta)}{(1-\eta)^2+(b/a)(\frac{1}{2}-\frac{2}{3}\eta)}, \tag{3}$$

and

$$\left(\frac{\mathrm{ReF}}{\mathrm{ReB}}\right)_{\mathrm{sphere}}=\frac{1+\frac{3}{2}\eta+\frac{1}{3}\eta^2+(b/a)[\frac{1}{3}+\frac{1}{4}\eta+(1/15)\eta^2]}{1-\frac{3}{2}\eta+\frac{1}{3}\eta^2+(b/a)[\frac{1}{3}-\frac{1}{4}\eta+(1/15)\eta^2]}, \tag{4}$$

respectively.

Equations (3) and (4) may be used for normalization provided that one has evidence that the center-of-mass distribution is of the form $a+b\cos^2\theta'$, and that η and b/a are known. The first provision can be satisfied by inspection of the measured (laboratory) angular distributions shown in Figs. 2, 3, and 4. Even before the forward and backward points are normalized to each other, the general shapes indicate a forward motion of the fissioning nucleus superimposed upon an anisotropic center-of-mass distribution. (If the center-of-mass distribution were isotropic, the observed cross section would continue to decrease beyond $\theta=90°$ with a slope comparable to that of the forward points.) The only center-of-mass distribution function capable of raising the observed cross section in the backward direction is one in which even powers of $\cos\theta'$ predominate. One is then justified in analyzing the data in terms of a center-of-mass distribution of the form $a+b\cos^2\theta'+c\cos^4\theta'+\cdots$. Since the anisotropies are small, the calculations have been limited to the determination of the single parameter b/a, no attempt having been made to determine coefficients of possible higher order terms.

An inspection of Eqs. (3) and (4) shows that the effect of the b/a terms on the normalizing factors is small, especially since b/a itself is shown by subsequent analysis to be small. The values of η obtained from the stacked-foil experiments and Eq. (1), therefore, are changed very slightly by a correction for center-of-mass anisotropy. The values of η from the stacked-foil experiments may consequently be used in calculating the center-of-mass distributions from the laboratory distributions. If it is assumed that the momentum vector of the fissioning nucleus is directed along the proton axis, it can be shown that, for a spherical target, the relationship between the laboratory and center-of-mass distributions is given by

$$\frac{\mathrm{Re}(\theta_1,\theta_2)}{\langle\sigma'(\theta_1',\theta_2')\rangle}=\frac{k}{3\eta}\left[(1+\eta^2+2\eta\cos\theta')^{\frac{3}{2}}\right]_{\theta_2}^{\theta_1}, \tag{5}$$

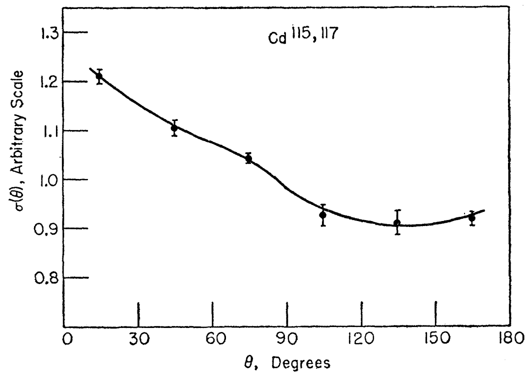

FIG. 4. Angular distribution of cadmium fragments in the laboratory system.

where $\mathrm{Re}(\theta_1,\theta_2)$ is the number of recoils observed in the angular interval between θ_1 and θ_2, $\langle\sigma'(\theta_1',\theta_2')\rangle$ is the average value of the center-of-mass cross section in the angular interval between θ_1' and θ_2' (corresponding to θ_1 and θ_2), and k is a constant involving the source radius and a range-energy proportionality constant. The laboratory and center-of-mass angles are related by the equation

$$\cos\theta'=\cos\theta(1-\eta^2\sin^2\theta)^{\frac{1}{2}}-\eta\sin^2\theta, \tag{6}$$

in which the radical approximates unity, since η is small.

The angular distributions in the center-of-mass system were obtained from the laboratory distributions by the use of Eqs. (5) and (6). A correction was then made for the fact that, in determining η from the stacked-foil experiments, an isotropic center-of-mass distribution had been assumed. The correction was accomplished by fitting the center-of-mass distribution by least squares to a function of the form $a+b\cos^2\theta'$ and using the resultant value of b/a to renormalize the experimental points and calculate a new η by means of Eqs. (3) and (4). The new η was then used to calculate a new center-of-mass distribution, etc. This successive approximation procedure converges rapidly, and only one iteration was required to determine the center-of-mass distributions to the accuracy justified by the experimental precision.

IV. RESULTS AND DISCUSSION

A. Data

The data from a typical forward angular-distribution experiment are given in Table I. The numbers in the next to the last column are the fractions of total forward activity (ReF) found in the three angular cuts 0°–30°, 30°–60°, and 60°–90°. When these numbers are divided by the fractional solid angles (0.134, 0.366, and 0.500, respectively), the numbers in the last column result. The latter are proportional to $\sigma(\theta)$, which is the number of recoils emitted at the laboratory angle θ per unit of

TABLE I. Data from a typical forward angular-distribution experiment.

Fission product separated	Laboratory angle, degrees	Activity corrected for chemical yield and self-absorption, counts/min	Activity corrected for blank, counts/min	Fractional activity $\left(\frac{\text{activity}}{\text{ReF}}\right)$	$\left(\frac{\text{Fractional activity}}{\text{fractional solid angle}}\right)$
$Ga^{72,73}$	0°–30°	212	203	0.155	1.157
	30°–60°	497	482	0.368	1.005
	60°–90°	637	626	0.477	0.954
	ReF	1346	1311		
$Sr^{91,92}$	0°–30°	440	440[a]	0.152	1.134
	30°–60°	1048	1048	0.363	0.992
	60°–90°	1400	1400	0.485	0.970
	ReF	2888	2888		
$Cd^{115,117}$	0°–30°	130	103	0.154	1.149
	30°–60°	285	244	0.366	1.000
	60°–90°	354	321	0.481	0.962
	ReF	769	668		

[a] Impurity activation leading to the production of Sr^{91} and Sr^{92} was negligible.

solid angle. The average normalized values of $\sigma(\theta)$ are plotted *versus* θ in Figs. 2, 3, and 4. The center-of-mass angular distributions are shown in Figs. 5, 6, and 7, fitted by the method of least squares to functions of the form $a+b\cos^2\theta'$. In each case the root-mean-square deviation of the points from the fitted curve is about 2% of the average cross section over the entire range of θ'.

Table II summarizes the anisotropy parameters (b/a) and center-of-mass motions (η) obtained from the data by the methods outlined in Sec. III above, which are based on the assumption that the center-of-mass motion is in the direction of the incident proton. The value of η for $Sr^{91,92}$ in Table II is slightly higher than the value 0.045 ± 0.005 reported for Sr^{91} by Sugarman, Campos, and Wielgoz,[13] who used a stacked-foil technique and assumed isotropy in the center-of-mass system. Correction of the stacked-foil η for an $a+b\cos^2\theta'$ anisotropy with $b/a=0.115$ gives good agreement, however, with the present value.

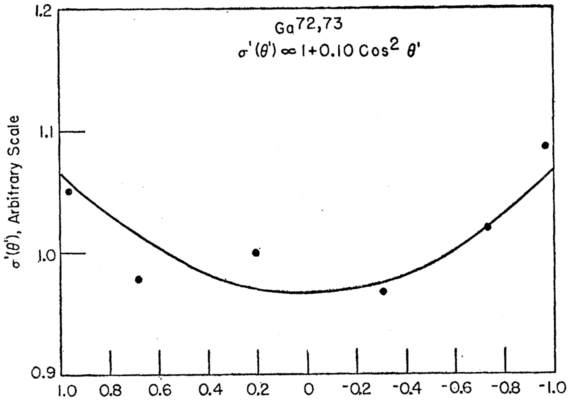

FIG. 5. Angular distribution of gallium fragments in the center-of-mass system.

The errors in the values of b/a shown in Table II are standard errors representing the goodness of least-squares fit of an $a+b\cos^2\theta'$ function to the calculated center-of-mass points. The errors in the points themselves are comparatively small.

The anisotropy parameters found in the present work for 450-Mev proton fission of Bi are of the same magnitude as those found at much lower energies (~20 Mev) for the proton-[4,5] and neutron-induced[1,6,7] fission of Th and U. For the symmetric fragments of the 22-Mev proton fission of U^{233}, U^{238}, U^{235}, and Th^{232}, Cohen *et al.*[5] have found $a+b\cos^2\theta$ distributions with b/a values of 0.07 to 0.10. In the 450-Mev proton fission of Bi, we find the symmetric fragment to have a b/a of 0.115. As the fragment asymmetry is increased, however, there is a significant difference in behavior between the 22-Mev and the 450-Mev processes. At 22 Mev, the fragments are emitted with increasing anisotropy as the mass ratio is increased. For Th^{232}, for example, b/a increases by a factor of about 2.5 as the mass ratio increases from 1.0 to 1.7[5]; the dependence is less pronounced for the uranium isotopes. For 450-Mev protons on bismuth, however, we find b/a to be practically constant with mass ratio.

B. Discussion

As noted earlier in Sec. III, the foregoing treatment is based on a model in which the fissioning nucleus,

TABLE II. Anisotropies and center-of-mass motions.

Fission fragment	Approximate mass ratio[a]	η	b/a
$Ga^{72,73}$	1.6	0.057 ± 0.003	0.10 ± 0.01
$Sr^{91,92}$	1.0	0.050 ± 0.002	0.115 ± 0.015
$Cd^{115,117}$	1.7	0.053 ± 0.004	0.09 ± 0.01

[a] Assuming a fissioning nucleus of mass 186 [P. Kruger and N. Sugarman, Phys. Rev. 99, 1459 (1955)].

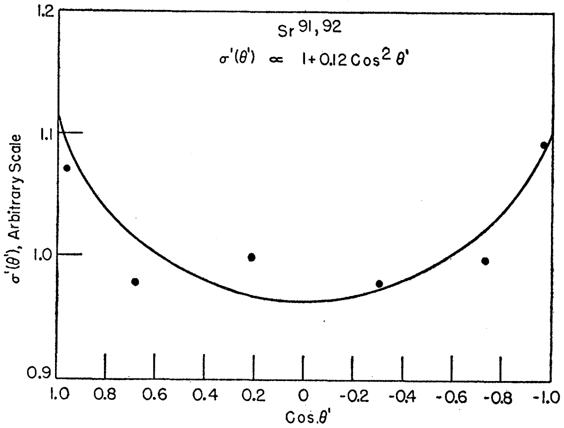

FIG. 6. Angular distribution of strontium fragments in the center-of-mass system.

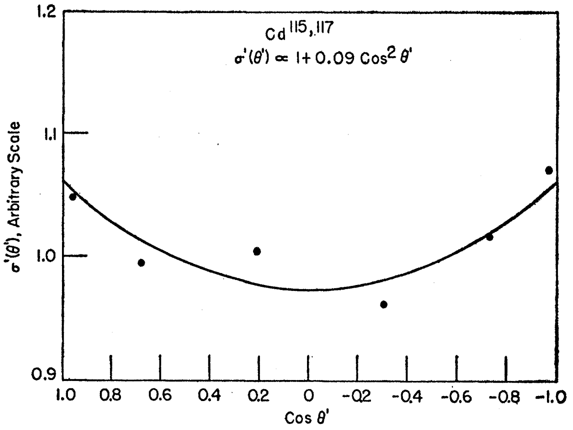

FIG. 7. Angular distribution of cadmium fragments in the center-of-mass system.

having acquired momentum from the proton, is assumed to be traveling in the direction of the incident proton when it fissions. It is interesting to consider the consequences should this assumption be invalid: that is, if there exists on the average a finite angle θ_0 between the incident proton direction ($\theta=0$) and the direction of the center-of-mass motion. We may consider, then, an alternate model in which the heavy nucleus, after being excited by the proton, is moving at an angle θ_0 to the proton axis when it fissions. (In the present discussion it is immaterial whether nucleon "evaporation" takes place before or after the fission act.)

Since θ_0 determines a preferred direction of motion from which the fission fragments are projected, one observable effect of θ_0 should be an enhanced cross section in some preferred direction in the laboratory system. Furthermore, since θ_0 must be smaller than $\pi/2$, this enhancement should be observed somewhere in the forward hemisphere. The bumps between 60 and 90 degrees in the laboratory angular distributions of Figs. 2, 3, and 4, then, may be an indication that θ_0 is not zero. These bumps persist in the derived center-of-mass distributions (Figs. 5, 6, and 7) when the data are treated by the present methods, in which it is assumed that $\theta_0=0$. Inasmuch as the bumps occur in all three of the measured angular distributions, the possibility of a systematic experimental error must, of course, be considered. In view of the fact, however, that the experimental arrangements were identical in the forward and backward runs except for the orientation of the cone with respect to the beam, it is unlikely that a systematic error could have the effect of raising the 60°–90° point without also affecting the 90°–120° point.

Apparently, then, the lack of perfect fit to an $a+b\cos^2\theta'$ function in the forward direction is a direct result of applying the $\theta_0=0$ model to a real effect shown by the experimental data. Two possibilities exist: either the $\theta_0=0$ model does not apply, or the center-of-mass distributions cannot adequately be represented by as simple a function as $a+b\cos^2\theta'$. If the latter is the case, a more sensitive experiment than the present one is necessary to determine what other terms must be added. If the model is at fault, it has been pointed out above that the most promising alternate model is that in which the $\theta_0=0$ assumption is dropped. A model in which $\theta_0\neq 0$ must be calculated in order to determine whether the data are in better agreement with such a model than with the assumption that $\theta_0=0$. Should the $\theta_0\neq 0$ model be the more successful one, the derived quantities η and b/a in Table II would change somewhat in magnitude. It is unlikely, however, that the dependence of b/a on mass ratio would be affected.

V. ACKNOWLEDGMENTS

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