Anisotropic Photofission*

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The angular distribution of photofission fragments with respect to the direction of a high-energy x-ray beam has been found to be anisotropic. The distribution seems to be of the same form, $a+b\sin^2\theta$, in a large variety of situations. The ratio of b to a depends on the energy of the photons producing the fission, on the particular fissionable target being irradiated, and on the particular fission fragments being observed. The photofission excitation curve is compared with the (γ,n) excitation curve in the energy region near the fission threshold where the fission anisotropy seems to be largest. Recent attempts to develop a theory of anisotropic fission are briefly discussed.

I. INTRODUCTION

IT was found several years ago that when high-energy x-rays cause nuclei to fission, the fission fragments are not emitted isotropically. More fragments are emitted at 90° to the x-ray beam than either forward or backward. This anisotropy was an unexpected result and a variety of aspects of the phenomenon have been studied in an effort to understand it. The size of the anisotropy has been examined as a function of photon energy for a number of fissionable nuclides. The dependence of the anisotropy on the mass ratio of the fission fragment pair has been studied, and some features of the relation between photoneutron emission and anisotropic photofission have been investigated. These experiments were all performed on the 16-Mev linear electron accelerator at M.I.T. and some of the results have already been reported briefly.[1–3] This paper contains an account of all of the experimental results obtained so far. It should be mentioned that fission produced by neutrons,[4] protons,[5] and α particles[6] has also been found to be anisotropic. It is therefore reasonable to infer that the particular method of exciting a nucleus to fission is of somewhat secondary importance as regards anisotropic fission and that the essential features very probably have to do with the nature of the fission process itself.

Although there have recently been some developments in the theory of fission in which attempts are made to account for the anisotropy of fission,[7,8] the experiments to be described were performed before any such theoretical guidance was available. It is for this reason that the most connected account of them is probably a chronological one. The experiments on anisotropic photofission are described in the following sections in the order in which they were done.

II. ANGULAR DISTRIBUTION OF PHOTOFISSION FRAGMENTS FROM THORIUM

The study of the angular distribution of photofission fragments was motivated in part by the explanation given by Goldhaber and Teller[9] of the giant resonance for photon absorption by nuclei. They suggested that the neutrons in a nucleus can oscillate more or less as a unit against the protons. Reasonable estimates of the natural frequency of this dipole oscillation agree fairly well with the observed frequency of the photons absorbed in the giant resonance. This explanation was reminiscent of another situation in which a simple hydrodynamical model of the nucleus had been found to be useful, namely in fission. To be sure, the liquid drop model of fission deals with a nuclear fluid in which there is no separation of charge.[10] This makes it very different from the Goldhaber-Teller model and yet it is possible to imagine a connection. The dipole oscillation excited in a nucleus by a high-frequency electromagnetic wave could perhaps persist in its original direction even after the neutrons and protons have once again been homogenized. This direction is along the electric vector of the incoming wave; that is, perpendicular to the x-ray beam. If the oscillation happens to lead to fission, the fragments would tend to come off at right angles to the beam.

The first attempt to observe the angular distribution of fission fragments was made with thorium because it was readily available. The x-ray beam was produced in a thick lead target by an electron beam whose spectrum was centered at 13 Mev and was about 5 Mev wide. The x-rays were passed through a thin cylindrical shell of thorium surrounded by a concentric plastic cylinder [see Fig. 1(a)]. The fission fragments emitted from the thorium were caught in the plastic and their

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1 Winhold, Demos, and Halpern, Phys. Rev. **85**, 728(A) (1952).
2 Winhold, Demos, and Halpern, Phys. Rev. **87**, 1139 (1952).
3 Fairhall, Halpern, and Winhold, Phys. Rev. **94**, 733 (1954).
4 J. E. Brolley, Jr., and W. C. Dickinson, Phys. Rev. **94**, 640 (1954); Brolley, Dickinson, and Henkel, Phys. Rev. **99**, 159 (1955).
5 Cohen, Jones, McCormick, and Ferrell, Phys. Rev. **94**, 625 (1954); R. L. Wolke, Phys. Rev. **98**, 1199 (1955).
6 C. T. Coffin and I. Halpern (to be published).
7 D. L. Hill and J. A. Wheeler, Phys. Rev. **89**, 1102 (1953).
8 A. Bohr, *International Conference on the Peaceful Uses of Atomic Energy*, Geneva, 1955 (United Nations, New York, 1956), Vol. 2.
9 M. Goldhaber and E. Teller, Phys. Rev. **74**, 1046 (1948).
10 N. Bohr and J. A. Wheeler, Phys. Rev. **55**, 426 (1939).

angular distribution was determined from a measurement of the distribution of β activity in the plastic. For this purpose a Geiger counter and slotted shield were used [Fig. 1(b)]. It was established that neither fast neutron fission nor the photon activation of the plastic contributed observably to the measured activities. Small corrections for the variation of x-ray flux over the target and for the angular resolution of the exposure and counting setups had to be made. The corrected data, based on a number of runs, is shown in Fig. 2.

The observed angular distribution is seen to be peaked at 90° to the beam and, within the errors of the data, it is symmetrical about this angle. The form of the distribution is compatible with $a+b\sin^2\theta$. If one fits the data of Fig. 2 with a curve of this form, it is found that $b/a=0.41\pm0.05$.

It should be emphasized that the actual measurement was one of the distribution of fragment *activities* rather than of the fragments themselves. If different species of fragments have different angular distributions, the observed activity distribution might be expected to depend on the length of the exposure and on the time that elapses between exposure and counting. It was found, however, that b/a showed no observable dependence on the exposure duration (which varied from 6 min to 3 hours) nor on the time of counting (up to several days after exposure). Evidence discussed in Sec. VII indicates that the fragment assortments are nevertheless somewhat different at different angles. These differences are apparently not great enough to be observable in the decay curves observed at the different angles.

III. ENERGY DEPENDENCE OF ANISOTROPIC PHOTOFISSION IN THORIUM

Angular distributions of reaction products following an electric dipole absorption would have to be of the form $a+b\sin^2\theta$. The observed distribution was therefore consistent with the original supposition that it was connected with the electric dipole absorption in the giant resonance.

To check this possible connection somewhat further, rough measurements were made of the anisotropy as a function of photon energy.[2] The giant resonance in thorium is centered at about 14 Mev[11] and the linear accelerator was able to accelerate electrons to about this energy. From observations of b/a as a function of electron energy, together with knowledge of the excitation curve for photofission and the shape of the x-ray spectrum, one can in principle extract b/a as a function of photon energy. There are fairly large uncertainties in the various components of the calculation, but fortunately the dependence of b/a on electron energy turned out to be so extreme that the qualitative dependence of b/a on photon energy was clear. The photons in the giant resonance region were found to produce essentially isotropic fission. Indeed, the observations were consistent with the assumption that the anisotropic fission is due solely to photons within about 3 Mev of the fission threshold. (See Table I.) More precise data confirming this general conclusion are discussed toward the end of the next section.

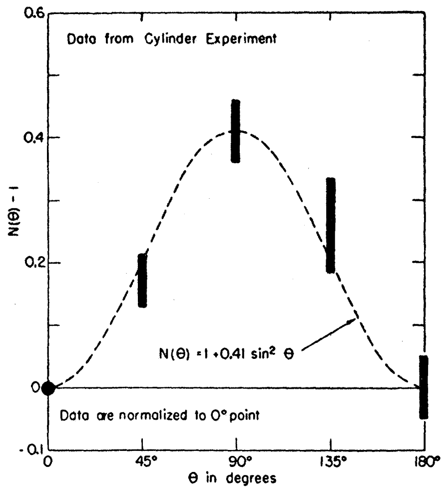

FIG. 2. The angular distribution, $N(\theta)$, of fission fragments from Th^{232} caught at the angle θ to the x-ray beam. The distribution was obtained with the equipment of Fig. 1 and with an x-ray spectrum where the maximum energy was 16 Mev.

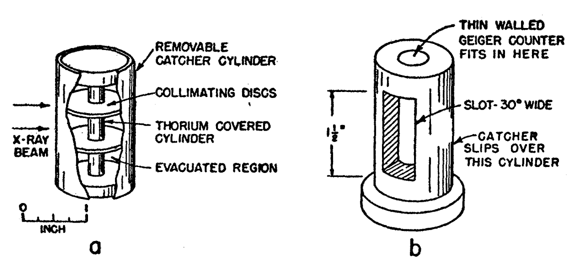

FIG. 1. The arrangements for exposing and counting the cylindrical fragment catcher. After exposure, the catcher is slipped over the cylinder at the right and oriented to permit the counter inside to "see" the activity caught at any desired angle to the x-ray beam.

IV. ANISOTROPIC PHOTOFISSION IN DIFFERENT TARGETS

The fact that the photons responsible for anisotropic fission are not the giant resonance photons makes it very doubtful that there is any connection after all between fission and the Goldhaber-Teller oscillation.

Yet the form of the observed distribution is suggestive of electric dipole absorption. Such an absorption would give rise to excited states in thorium in which the angular momentum of the nucleus, J, is equal to 1 with $m=\pm1$ (since $J=0$ in the thorium ground state). The observed angular distribution consists of an isotropic part plus a part where the orbital angular momentum, L, between fragments is likely 1 and $m_L=\pm1$. A large anisotropy could be expected if there

[11] R. Nathans and J. Halpern, Phys. Rev. 93, 437 (1954).

is, for some reason, a tendency for the thorium nucleus to break up with its angular momentum going into the orbital motion of the fragments rather than into their internal spins.

One might reasonably wonder whether there is any need to assume special restrictions on the distribution of nuclear angular momentum in fission in order to account for the observed anisotropy. After all, the excited nuclei have $J=1$ and are lined up with their spins along the beam axis. Therefore since $L+J'=J$ (where J' is the vector sum of the spin angular momenta of the two fission fragments) both L and J' will *on the average* also be lined up along the beam when no special assumptions are made about the apportionment of angular momentum during fission.

One can show, however, that the amount of lining up that could be expected on purely statistical grounds is very small indeed. For example the $L=1$ breakups would be strictly isotropic if the probability of a breakup is proportional to $2J'+1$. If one assumes instead that this probability is independent of J', the $L=1$ breakups lead to the distribution $1+\frac{3}{16}\sin^2\theta$. If to this distribution is added the isotropic one arising from $L=0$ emissions, it is seen that there would be hardly any anisotropy at all. It is clear, on the other hand, that if one were to make the extreme assumption that J' is always zero, there would be plenty of anisotropy. That is, if at the critical time in the fission process when final angular momenta are being determined, there is some reason to favor the assignment of zero spin or very low spin to each of the fragments, then one might expect anisotropy on a statistical model. However, it is hard to see under what circumstances it would be reasonable to expect the required suppression of internal spins of the fragments. Such a suppression could perhaps be connected to the energetics of the fission process. One might suppose that those nuclei are most likely to fission, which, in the course of their oscillations, approach the saddle point distortion[10] with their nucleons in the lowest possible energy states. The lowest energy configurations might involve an extreme pairing of nucleon spins. Such a view of the nature of anisotropic photofission would not be inconsistent with the observed rapid disappearance of the anisotropy as the excitation energy is raised beyond a few Mev above the threshold. Although this "explanation" of anisotropic fission is essentially statistical in its viewpoint and deals only with energies and spins, it requires an important additional assumption about the behavior of a fissioning nucleus; for example, that spins of nucleons show a very strong tendency to pair. The basis for such an assumption would have to be sought in terms of special restrictions on the dynamical properties of nuclei, that is, in terms of some particular "nuclear model." The idea of nuclear spin pairing at the saddle point has recently been developed by Bohr in terms of the collective model,[8] and is discussed in Sec. VIII.

These considerations about the possible connection between the anisotropy and nuclear spins made it seem very desirable to look at fragment angular distributions from some nuclide having nonzero spin in its ground state. It was decided to examine U^{235} where the ground state spin is 5/2 or 7/2.[12] An electric dipole absorption would lead to states with assorted high values of J and the "directional content" of the excited state would be less than that of the corresponding thorium state. The fission fragment distribution would be expected to be much less anisotropic even near the threshold.

By the time that we were able to obtain some U^{235}, we had managed to increase the intensity of the electron beam of the linear accelerator several-fold. The additional intensity permitted the use of a magnetically analyzed beam ($\sim 3\%$ line width). In addition it was possible, by a method to be described, to use thin-target x-radiation in the fission experiments with the result that the shape of the x-ray spectrum was much better known than it had been in the thick-target experiments. These improvements in technique permitted the conversion of observed dependences on electron energy to dependences on photon energy with much more reliability than had been possible before.

The ratio of the activity at 90° to the beam to the activity at 0° or 180° was measured for U^{235}, U^{238}, and again for Th^{232} by the apparatus illustrated in Fig. 3. The targets were all in foil form,[13] somewhat oxidized and thick to fission fragments (about 100 mg/cm^2). Target foils were placed at the center of the evacuated scattering chamber and oriented at 45° to the analyzed electron beam. The forward-directed high-energy x-rays produced by the electrons passing through the foil interacted with the fissionable nuclei in the foil much more effectively than the electrons themselves. The thickness of each of the fission foils used was such that approximately the same number of fissions were being produced by x-rays and electrons at the back side of the

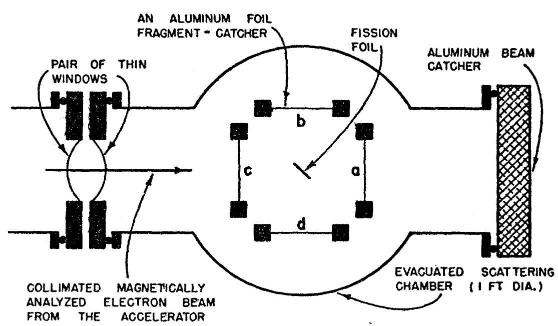

FIG. 3. Apparatus for measuring the anisotropy in photofission due to thin-target x-ray spectra.

[12] G. L. Stukenbrocker and J. R. McNally, Atomic Energy Commission Report AECD 2797 (unpublished).

[13] The isotopic purity of the U^{235} was about 90%. We are indebted to the U. S. Atomic Energy Commission for the loan of this foil.

foil. Since the range of fission fragments is much smaller than such a thickness, the number of fission fragments leaving the back side was *about twice as large* as the number of fragments emitted from the "entrance" side. Aside from small corrections due to electron energy loss in the fission foil, the 90°/0° activity ratio for fissions due only to the x-rays made in the fission foil is

$$(A_b - A_d)/(A_a - A_c).$$

The A's are the activities measured in the four catcher foils of Fig. 3, and it is assumed that the activity at 180° is identical to that at 0° (see Fig. 2).

A typical run lasted for one hour with an analyzed electron beam of 0.2 μa. After exposure, the catcher foils were rolled into cylinders and counted on thin-walled Geiger tubes. Tests were made which showed that the neutron-induced fission in U^{235} was negligible and that the multiple scattering of the fission fragments in the target did not significantly smear out the angular distribution. It was possible to estimate roughly the latter effect from the literature on the stopping and scattering of fission fragments,[14–16] but in addition a rough experiment was performed where fission fragments were scattered in thin gold foils.

Assuming that the angular distributions are all of the form $a+b\sin^2\theta$, the observed distributions were corrected for the finite angular resolution. The dependence of b/a on electron energy is shown for the three targets in Fig. 4. It is seen that the anisotropies in Th^{232} and U^{238} decrease rapidly with increasing electron energy. The most striking result is the absence (within experimental errors) of any anisotropy at all in U^{235}.

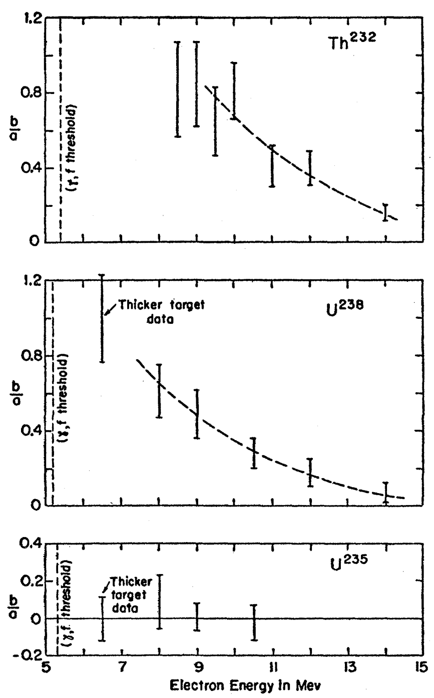

FIG. 4. The anisotropies in the photofission of three targets observed with the apparatus of Fig. 3. The angular distributions were all assumed to be of the form $a+b\sin^2\theta$.

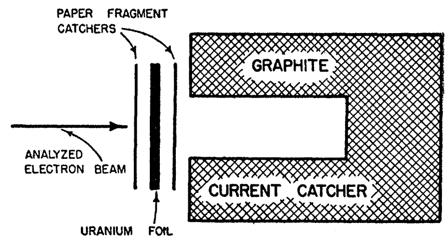

FIG. 5. The experimental setup used in the determination of the fission yield curve in U^{238} for thin-target bremsstrahlung.

It should perhaps be mentioned here that, with the help of measured photofission excitation curves (Sec. V), it was possible to plot the anisotropy as a function of photon energy for both Th^{232} and U^{238}. The qualitative thick target results of Sec. III were confirmed. The anisotropy drops to a small value at photon energies one or two Mev above the fission threshold. In the region of the giant resonance, the anisotropy is very small if not zero.

V. CROSS SECTION FOR PHOTOFISSION AS A FUNCTION OF ENERGY

It has been mentioned that in order to be able to re-express the data of Fig. 4 in terms of photon energy rather than electron energy, it was necessary to obtain the shapes of the photofission excitation curves. Although some studies of the dependence of photofission yield on energy had been made,[17–19] not enough information was available about the shapes of excitation curves at those photon energies that were proving to be significant in these experiments.

It was therefore decided to measure the fission yields for thin-target bremsstrahlung from threshold on up as far as we could conveniently go (about 12 Mev). Only U^{238} was examined directly. The yields of U^{235} and Th^{232} were obtained relative to the U^{238} yield. Such relative measurements are likely to be somewhat more sensitive to slight differences in yield curves than a comparison of independently measured curves.

The arrangement used in obtaining the yield curve for U^{238} is shown in Fig. 5. The magnetically analyzed electron beam was allowed to pass through a sandwich consisting of three foils in the sequence, catcher: uranium: catcher, and then into a large carbon Faraday cup. The difference-activity in the catcher foils per

[14] Bøggild, Brostrøm, and Lauritsen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, No. 4 (1940).
[15] N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **18**, No. 8 (1948).
[16] N. O. Lassen, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. **25**, No. 11 (1949).
[17] J. McElhinney and W. E. Ogle, Phys. Rev. **81**, 342 (1951).
[18] Huizenga, Gindler, and Duffield, Phys. Rev. **95**, 1009 (1954).
[19] R. B. Duffield and J. R. Huizenga, Phys. Rev. **89**, 1042 (1953).

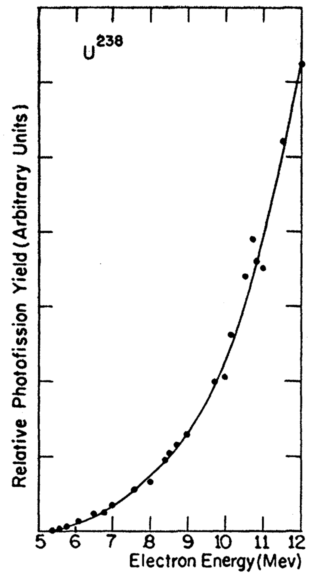

FIG. 6. The photofission yield in U^{238} as a function of electron energy. The data were obtained with the arrangement of Fig. 5.

unit of collected charge was obtained as a function of electron energy. Aside from some small corrections for electron loss in the uranium, the difference activity is a measure of the activity induced by thin-target bremsstrahlung. The data are plotted in Fig. 6. The ordinate, which is proportional to the measured fission activity, has been labeled "fission yield." This is justifiable because the activity per emitted fission fragment is quite independent of photon energy in the energy region investigated. More specifically, the assortment of fragments (the mass-yield curve) changes slowly with energy up to 12 Mev.[20,21]

Fission yields in U^{235} and Th^{232} were obtained relative to the U^{238} yield as a function of energy by bombarding a set of three sandwiches simultaneously. For example, to compare the yield in U^{238} with that in Th^{232} the beam was allowed to pass through the sequence of sandwiches, thorium: uranium: thorium. The average difference-activity in the thorium sandwiches was compared to the difference activity in the uranium. The yields of U^{235} and U^{238} have been plotted in terms of the yield in Th^{232} in Fig. 7. The relative yields are not nearly as energy-independent as they were found to be in the giant resonance (12–22 Mev) region.[17,18] This implies that when the fission cross-section curves for the three targets investigated are plotted as a function of photon energy, they must have different shapes at the lower energies. In order to obtain the excitation curves from the yield curves, the standard photon difference method was used with a Bethe-Heitler bremsstrahlung spectrum.[22] The most interesting feature of the excitation curves (Fig. 8) is the bump near the threshold that is apparently present in Th^{232} and to a lesser extent in U^{238} but not in U^{235}. There is always some uncertainty in obtaining excitation curves (cross section *vs* photon energy) from yield curves (yield *vs* electron energy). The procedure is essentially a differentiation and requires rather precise integral (yield) data in order to be meaningful. As an exercise, a number of different shapes of cross-section curves (i.e., curves where the abscissa is photon energy) were assumed and expected yield curves were computed. Only cross sections showing a rather extreme bump gave rise to yield curves that looked reasonably like that experimentally observed for Th^{232}. Another more precise fission excitation curve for Th^{232} was obtained later by another technique (see Sec. VI). It is essentially the same as that in Fig. 8. No attempt was made to measure any of the cross sections absolutely, but normalizing at the high-energy end of the present data to the data of Duffield and Huizenga[19] and to that of Katz *et al.*,[21] the cross section at the bump in U^{238} is roughly 12 mb. This is consistent with the measurement by Hartley[23] with monochromatic γ rays of 6.1 Mev. Using the photons emitted in the $F^{19}(p,\alpha\gamma)$ reaction, he finds a fission cross section for Th^{232} of 6 ± 2 mb.

The presence of a bump in the cross section of those targets that show an anisotropy and the absence of a bump for the target whose fragments are isotropic suggest a possible connection between the bump and anisotropic fission. This apparent connection is made to seem very reasonable by the observation (Sec. III) that anisotropic fission predominates at those photon energies which correspond to the location of the bumps.

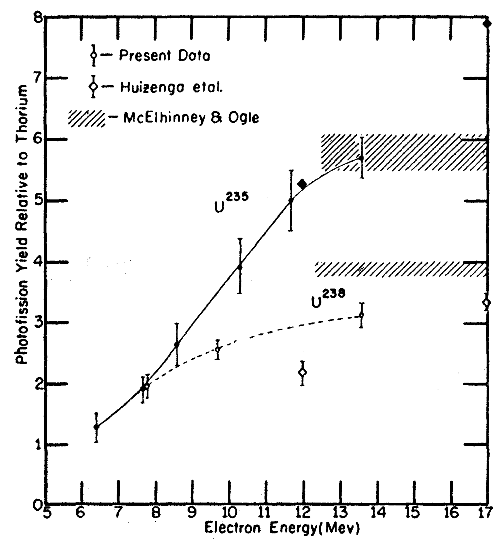

FIG. 7. The fission yields of U^{235} and U^{238} relative to the yield in Th^{232}. The data of Huizenga *et al.* are from reference 18 and those of McElhinney and Ogle from reference 17.

[20] R. A. Schmitt and N. Sugarman, Phys. Rev. 89, 1155 (1953); H. G. Richter and C. D. Coryell, Phys. Rev. 95, 1550 (1954).

[21] Katz, Kavanagh, Cameron, Bailey, and Spinks, Phys. Rev. 99, 98 (1955).

[22] See, for example, L. Katz and A. G. W. Cameron, Can J. Phys. 29, 518 (1951).

[23] W. J. Hartley, Ph.D. thesis, University of Pennsylvania, 1955 (unpublished).

Indeed, the data in the measurements described are altogether consistent with the assumption that anisotropic fission is confined to the region of the bump.

Two views of the meaning of the bump suggest themselves. In the first, it is assumed that there is a special mechanism for photon absorption that exhibits a resonance at about 6 Mev in some nuclides but not in others. One would further suppose that the state (or states) excited in the resonance have special properties which are responsible for the observed fission anisotropy.

In the other view, there is no particular connection between the bump and the anisotropy. The shapes of low-energy photofission excitation curves are held to depend on the nature of the competition between fission and neutron emission. This in turn depends on the relative locations of the thresholds for fission and neutron emission. Thus in U^{235} (see Table I), the neutron threshold is sufficiently close to the fission threshold to suppress fission for the first few Mev above threshold. In U^{238} and Th^{232}, on the other hand, fission gets off to a good start and is then reduced somewhat at the neutron threshold, with both cross sections rising again as the giant resonance is approached. According to this picture, the explanation for the dependence of the anisotropy on the fissioning nuclide is unrelated to the excitation curve dependence, and is to be sought separately.

It was felt that the detailed examination of photoneutron and fission excitation curves for at least one nuclide might prove useful. Such measurements would show whether the "competition" explanation of the excitation curves still seemed reasonable when better data were available. Accordingly a study was undertaken of neutron emission and fission in thorium at low energies. These measurements and their results are described in the following section.

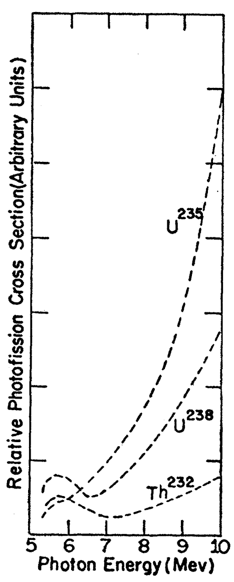

FIG. 8. The excitation curves for photofission obtained by the photon-difference method from the curves of Figs. 6 and 7.

TABLE I. Photofission and photoneutron thresholds.

Thresholds in Mev	(γ,f)	(γ,n)
Th^{232}	5.40 ± 0.22[a]	6.35 ± 0.10[b]
U^{235}	5.31 ± 0.25[a]	5.18 ± 0.17[b]
U^{238}	5.08 ± 0.15[a]	5.97 ± 0.10[b]

[a] Koch, McElhinney, and Gasteiger, Phys. Rev. **77**, 329 (1950).
[b] J. R. Huizenga, Physica **21**, 410 (1955).

VI. PHOTONEUTRON AND PHOTOFISSION CROSS SECTIONS IN THORIUM NEAR THE THRESHOLD

It was decided to compare fission and neutron emission in thorium, because, of the three targets studied, thorium had the most pronounced bump in the earlier measurements of fission excitation curves. It proved difficult to identify the (γ,n) activity in thorium radiochemically and it was therefore decided to count the neutrons emitted from the irradiated target.

The experimental arrangement is illustrated in Fig. 9. The analyzed electron beam was buried in a block of graphite inside an aluminum cup. The current collected by this target cup was measured and integrated. The x-rays produced were allowed to irradiate a thorium target. The spectrum of these x-rays is of course a thick-target spectrum. In order to obtain a thin-target spectrum, half the runs were made with a heavy metal foil a few mils thick in front of the graphite. The difference in observed counting rates with and without the heavy foil measures (aside from a few small corrections) the rate due to thin-target bremsstrahlung in the heavy metal foil.

The fission yields and neutron yields from thorium were measured simultaneously. The thorium target consisted of a few grams/cm^2 of thorium oxide in a thin but wide aluminum container onto the front and back of which were fastened a pair of thorium foils with their fragment-catcher foils. The thorium foils were as wide as the container of oxide and the activity on the associated catchers was taken as a measure of the fission yield for the flux of x-rays that passed through the container. The neutron yield was measured by the activity induced in a pair of rhodium foils placed at such a distance in the paraffin surrounding the whole setup that their neutron counting efficiency was expected to be reasonably independent of neutron energy. In order to check this aspect of the neutron detection, runs were made with a tantalum target at several energies from about an Mev above the photoneutron threshold (7.6 Mev) to about 14 Mev. The (γ,n) yield was measured both by rhodium detectors and by a measurement of the well-known activity of Ta^{180}. The relative yields obtained were identical within their fairly small errors.

The observed fission and neutron yields from thin target x-rays on thorium are shown in Fig. 10. The neutron yield generally includes both photoneutrons

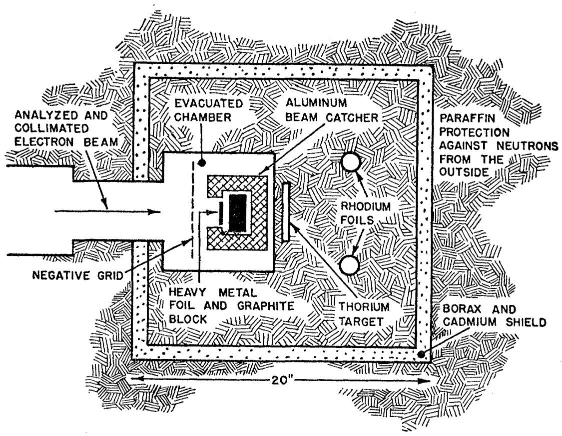

FIG. 9. The arrangement of apparatus for the comparison of the photofission cross section with the (γ,n) cross section in thorium.

and photofission neutrons, but in the energy region between the fission threshold (5.4 Mev) and the (γ,n) threshold (6.35 Mev) there should be only fission neutrons. As expected, the neutrons and fission yield curves are alike in this energy region. Above 6.35 Mev the neutron yield curve pulls away from the fission yield curve. In order to estimate the ratio of (γ,n) to (γ,f) yields in this region, it is necessary to know ν, the average number of neutrons emitted per fission. This number seems to be fairly independent of the type of projectile causing fission and of its energy, as long as this energy is not more than about 20 Mev.[24–26] Assuming that ν has the reasonable value of 2.5 throughout the energy region being examined,[27] one can obtain from the fission curve, the number of neutrons that must be subtracted from the neutron production curve in order to obtain the (γ,n) yield curve. Indeed, if the curve labeled "fission" in Fig. 10 is subtracted from the curve labeled "neutron production" and the difference curve is multiplied by 2.5, this new curve shows the number of (γ,n) events on the same scale as the fission curve.

It is possible to analyze the fission yield and the (γ,n) yield curves by the photon difference method to obtain relative cross sections as a function of photon energy. This has been done and the excitation curves are shown in Fig. 11. Although the scale for the ordinate is arbitrary, the relative cross sections for the (γ,n) reaction and for fission are presumably given correctly in the figure.

The very rapid rise of the photoneutron curve is probably quite real, although the details in the shapes of the curves in Fig. 11 are perhaps not to be trusted. The rapid rise is consistent with the idea that the drop in the fission curve is due to successful competition from neutron emission.

If the observed cross-section shapes are to be explained in terms of competition, it is to be expected that the sum of all reaction cross sections would give a reasonably smooth curve as a function of energy. The sum of the (γ,n) and (γ,f) curves is not smooth, but presumably the (γ,γ') curve drops enough at the neutron threshold so that the sum for all three cross sections does vary slowly with energy. Available evidence about sizes and shapes of (γ,γ') curves[28] is consistent with this assumption.

If one accepts the interpretation of the photofission excitation curve of the present discussion, he is admitting that low-energy photofission is "slow" rather than "direct". Thus photofission in the energy region of anisotropic fission would have to be slow enough so that neutron emission, which is usually assumed to be fairly slow, can successfully compete with it.

VII. DEPENDENCE OF THE ANISOTROPY ON THE FRAGMENT MASS RATIO

In this section, an aspect of anisotropic fission somewhat unrelated to the results of the preceding sections will be described. The anisotropy has been found to be strongly dependent on the mass ratio of the fragments. A preliminary account of this result has already been published.[3]

This experiment was originally undertaken in part because the counting of the total assortment of fission

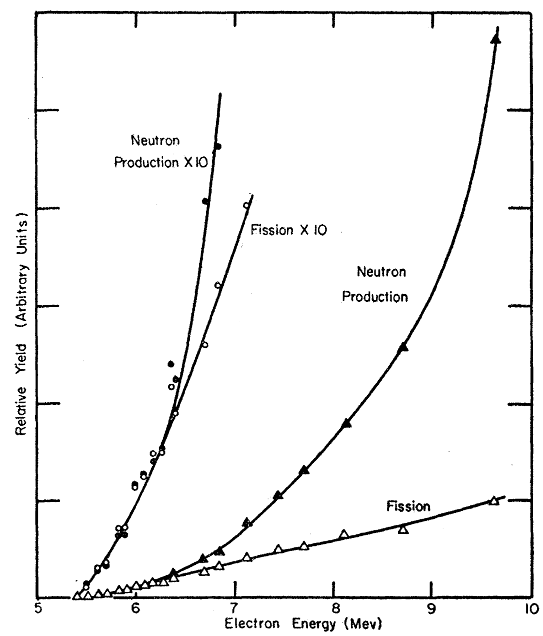

FIG. 10. Fission yields and neutron yields in Th^{232} obtained with the apparatus of Fig. 9. The curves have been normalized so that they fall on each other below 6.3 Mev (see text).

[24] E. Segrè, Phys. Rev. 86, 21 (1952).
[25] Barclay, Galbraith, and Whitehouse, Proc. Phys. Soc. (London) A65, 73 (1952).
[26] D. M. Hiller and D. S. Martin, Jr., Phys. Rev. 90, 581 (1953).
[27] "AEC declassified data," Nucleonics 10, No. 5, 64 (1952).
[28] Burkhardt, Winhold, and Dupree, Phys. Rev. 100, 199 (1955).

activities seemed to be unnecessarily crude. It also occurred to us that one might perhaps expect a connection between anisotropy and mass asymmetry of fission along the following lines. If the lighter fragments tend to come off with a different average value of Z/A from the heavier fragments, an asymmetric fragment pair would possess an electric dipole moment. Overlooking the expected fluctuations in the values of Z/A for the fragments in a given mass pair, the symmetric mass pair would have no dipole moment. If the observation of a moment between just-separated fragments is taken to indicate the existence of a corresponding moment within the target nucleus before fission (due, say, to a fluctuation in the charge distribution), then situations leading to asymmetric fission are more "ready" to absorb photons in dipole transitions. The angular distributions of the fragments due to these transitions would be of the observed form, namely $a+b\sin^2\theta$. Thus anisotropic fission is viewed here as being due to some fast or "direct" interaction, and one would think that it might be stronger for asymmetric fission than for symmetric fission. Actually, an examination of the best evidence[29] for the distributions of Z/A for final fission fragments shows too little difference between the various fragments to sustain the present argument. Moreover, we have seen that anisotropic fission is very likely a slow rather than a fast process.

Nevertheless, the experiment was performed and the correlation in question was found to exist.

The measurement was carried out with thorium and consisted of a comparison of the 90° yield to the average of the 180° and 0° yields for a number of specific fragments. The exposure arrangement involved a

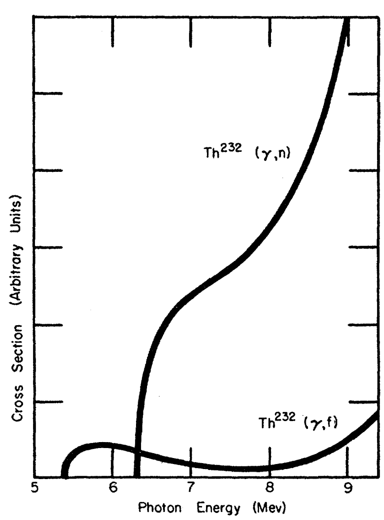

FIG. 11. Excitation curves for photofission and the (γ,n) reaction in thorium obtained from the yield curves of Fig. 10 (see text).

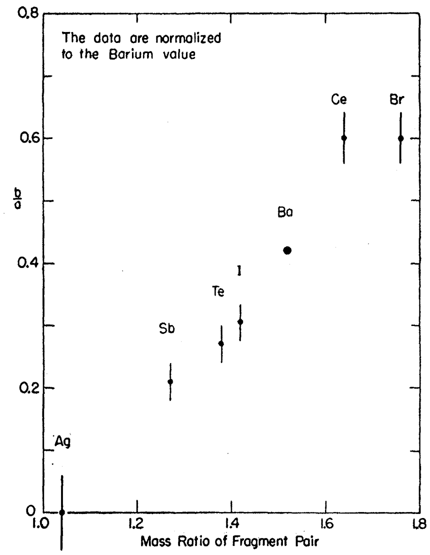

FIG. 12. The anisotropies in the distributions of various fragments in the photofission of thorium with thick-target x-rays of maximum energy 16 Mev. These data were obtained with foil stacks of the type shown in reference 3.

stack of target foils separated from their catcher foils by collimator disks. (An exploded view of a foil stack is shown in reference 3.) After exposure of the stack, the catchers were processed radiochemically. It was possible to obtain information on seven specific fission fragments. In each run, barium and several other elements were separated radiochemically from two stacks, one of which had been exposed lined up with the beam and one perpendicular to it. The data of Fig. 12 represent the results of many runs on each element. It has been assumed that all of the angular distributions are of the form $a+b\sin^2\theta$. The ordinates in Fig. 12 are the values of b/a normalized to that for 85 min Ba^{139}. The b/a value for Ba^{139} was obtained by a careful comparison with that for the unseparated activities. The ratio between these b/a values was found to be 1.00 ± 0.01. For reasons of intensity, the measurements being described were all performed with thick target 16-Mev bremsstrahlung. Under these conditions, b/a for the unseparated activities had been measured to be 0.41 ± 0.05 (see Sec. II).

It is seen that the anisotropy seems to increase monotonically, perhaps linearly, with the mass ratio, mass-symmetric fission being essentially isotropic.

It is perhaps instructive to think of the implications of Fig. 12 for the shapes of the mass distribution curves for isotropic and anisotropic fission. Assuming that the total mass distribution curve in thorium resembles that in uranium where it has been measured,[21] it is possible to construct mass distribution curves for the isotropic and anisotropic components of the angular distribution separately. These curves each have essentially the same familiar double-humped shape as their sum. The peaks of the anisotropic curves are slightly further apart than

[29] L. E. Glendenin, Technical Report No. 35, Laboratory of Nuclear Science and Engineering, Massachusetts Institute of Technology, 1949 (unpublished).

those of the isotropic curve owing to the correlation of Fig. 12, but the difference is small. It is probably worth emphasizing this point, lest one get the impression from Fig. 12 that the mass distribution for anisotropic fission is very abnormal. Those who view the double-humped mass distribution curve as evidence for the slowness of the fission process would have to conclude that anisotropic fission is no faster than other fission.

Similar correlations between anisotropy and mass ratio have been observed by Cohen and his co-workers[30] with 22-Mev protons. Here again, the more asymmetric mass pairs show the larger anisotropy although the character of the anisotropy is different. More fragments come out forward and backward than sideways.

It is probably unlikely that there is much to be gained at this time from a detailed comparison of correlation experiments of this type. There are certain ambiguities in each experiment that tend to interfere with comparisons. For example, the data for photons (Fig. 12) is for a 16-Mev thick-target x-ray spectrum. Thus the correlation in the figure is some sort of average correlation for photons up to 16 Mev. In view of the data of Fig. 4, it is reasonable to assume that the higher energy photons give rise to isotropic fission so that their contribution acts generally to reduce b/a throughout the graph. In particular, the silver yield is probably due mostly to photons above the energy connected with anisotropy. This is because the probability for fission into nearly symmetric masses increases very rapidly with photon energy.[21] If it were possible to obtain the correlation between anisotropy and mass distribution due only to those photons (6–7 Mev) giving rise to anisotropic fission, one would very likely find values of b/a larger than those in Fig. 12 for the more asymmetric fragments. One can only guess whether the small amount of symmetric fission that occurs would show any anisotropy.

Although the proton fission data, in contrast to the photon data, are due to protons of a unique energy, there are here too some features that make comparisons difficult. It is hard to know what role is played by the emission of neutrons before fission in determining both the mass distribution and angular distribution of fission fragments.[30]

Inasmuch as no generally accepted explanations of the fission mass distribution or angular distribution exist, it is not surprising that a correlation between them is difficult to explain. There have been some recent suggestions[8] that fissionable nuclei tend to be asymmetric in shape at low excitation energies, but it is probably too soon to make any serious attempts to account for the correlation described in this section.

VIII. SUMMARY OF RESULTS

In the preceding sections a number of experimental features of anisotropic photofission have been described. Stated, for simplicity, with rather more generality than the observations warrant, it has been found that:

(1) The form of the anisotropic angular distributions is $a+b\sin^2\theta$.

(2) Not all fissionable targets give anisotropic distributions.

(3) Where there is anisotropy, it appears only for those photons whose energies are within a couple of Mev of threshold.

(4) The dependence of the fission cross section on photon energy is different for targets that do fission anisotropically from those that do not.

(5) There is a correlation between the anisotropy and the mass ratio of the observed fission fragments.

In any attempt to account for these observations, one of the more basic questions to answer has to do with the speed of anisotropic photofission. One could look for explanations in terms of models involving either a relatively long-lived compound state or in terms of some fast "direct" interaction of the photons with a fissionable nucleus.

It would seem that photofission, including anisotropic fission, is a slow process. One of the reasons for this conclusion is the evidence (Sec. VI) that photoneutron emission competes successfully against photofission. It was seen that at the photoneutron thresholds in Th^{232} and U^{238}, the fission is depressed. This suggests that fission is at least as slow as photoneutron emission. Since observations of photoneutron spectra[31,32] at low energies seem to show that photoneutrons are evaporated from a compound nucleus, it follows that photofission must also involve the formation of a compound nucleus.

Another indication that anisotropic fission is slow is the fact that it occurs quite generally. It has been observed with all sorts of projectiles, and most of the observations have been made at bombarding energies at which the major part of the reaction cross section seems to involve the formation of a compound nucleus. Any theory of the anisotropy of fission based on some fast direct mechanism would have to explain how this mechanism operates in the large variety of situations in which such fission has been observed.

Although it would seem from the foregoing that anisotropic fission is slow, it was seen (Sec. IV) that purely statistical considerations on the breakup of a fissioning nucleus do not lead to sufficiently large anisotropies. Some special assumptions have to be made about the structure of the compound nucleus.

Considerable information about the structure and behavior of heavy nuclei has recently been obtained and correlated in terms of the so-called collective or unified model of the nucleus.

The first suggestion that anisotropic fission might

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

[31] G. A. Price, Phys. Rev. **93**, 1279 (1954).

[32] W. E. Stephens (private communication).

be accounted for on the basis of the collective model was made by Hill and Wheeler.[7] In their view, a single nucleon picks up the excitation energy from the bombarding agent and feeds it into nuclear surface oscillations. The probability of picking up this energy depends on the orientation of the nucleon's orbit with respect to the beam. But the direction of the distortions induced in the nuclear surface by a nucleon depend on the orientation of the nucleon's orbit with respect to the surface. In this way, it is possible for distortions or oscillations to be induced in a nuclear surface in some preferred direction to an incoming beam. Such preferentially oriented oscillations would lead to anisotropic fission.

Since no quantitative predictions were made by Hill and Wheeler, it is difficult to compare their ideas with the experimental observations. It would seem, however, that the mechanism suggested is rather "direct" and it is not clear how the competition of neutron evaporation with fission would be accounted for on the basis of this model. It should be mentioned that the Hill-Wheeler picture of anisotropic fission implies, in accord with observations, that most fragments are emitted sideways for photon-induced fission and forward and backward for particle-induced fission. This prediction is unfortunately not a very critical test of the theory. Any theory in which the orbital motion between the fission fragments is given a fair share of the angular momentum brought in by the bombarding agent, would give the same prediction.

More recently Bohr has developed a view of anisotropic fission based on the collective model that seems to account, at least qualitatively, for the observations listed at the beginning of this section.[8] It was indicated in Sec. IV that any compound nuclear theory of anisotropic fission would have to provide some plausible mechanism for concentrating the angular momentum absorbed from the beam on the orbital motion between the fragments. If the fragments are allowed to have appreciable angular momenta themselves, too many possibilities for the distribution of the angular momentum become available and the angular distributions tend to be isotropic.

In Bohr's view, the orbital motion of the fragments grows out of a collective rotation of the nucleus. He assumes that the rate of the distortion of the nucleus in fission is slow enough so that the nucleons tend to find themselves in the lowest possible states determined by the potential corresponding to the distortion. The quasi-equilibrium behavior is assumed to persist through the saddle point distortion. The order of the levels at the saddle point distortion is assumed to resemble the order near the ground state for unexcited heavy nuclei. There apparently exists a low-lying 1^- collective state in such nuclei for which the angular momentum vector is perpendicular to the nuclear symmetry axis. In photofission it is assumed that even-even nuclei like Th^{232} and U^{238} absorb dipole photons and that for energies near threshold they pass through the saddle in the 1^- state that corresponds to the state just mentioned. Since the rotation of the nucleus is perpendicular to the angular momentum in this state, the fragments tend to fly apart perpendicularly to this vector and hence perpendicularly to the x-ray beam. The essential idea is that the nucleus is "cold" enough at the saddle point so that the nucleon spin pairing that takes place for the lowest lying states in heavy nuclei also takes place at the saddle point.

For excitation energies even a few Mev above threshold, many other 1^- states become available at the saddle point distortion. In these states, the nuclear symmetry axis has various orientations with respect to the nuclear angular momentum, and the angular distribution tends to become isotropic. Anisotropic photofission is expected to be observable only within a few Mev of threshold.

In U^{235}, which has a large spin in its ground state, a dipole absorption leads to an excited nucleus whose angular momentum vector is almost isotropically oriented with respect to an x-ray beam. Moreover, odd-A nuclei are expected to have a greater concentration of levels near the saddle point in which the spin is carried by single nucleons instead of collective oscillations. For both these reasons, one is led to expect a rather isotropic distribution of fragments in the photofission of U^{235}. (See Fig. 4.)

Bohr finds it possible, on the basis of this model, to give a qualitative account of some of the observations in particle-induced fission as well as in photofission. For particles, neither the ground state spin of the target nor the excitation energy are expected to be quite so important as they are in photofission. This is because of the large amount of angular momentum brought in by bombarding particles especially at higher energies. Indeed, because of this momentum one can expect anisotropy in particle-induced fission at energies far above the fission threshold. Again it is the tendency for nucleon spins to pair that is important in determining the fragment angular distribution. The component of the nuclear angular momentum along the nuclear symmetry axis is due to the momentum associated with individual nucleons and tends to remain small. The angular momentum brought into the nucleus tends to feed into the collective rotational motion and since this momentum is oriented perpendicular to the beam, the rotations tend to take place in planes containing the beam. One is led in this way to expect particle-induced fission to give rise to fragments emitted mostly forward and backward and one is also led to expect anisotropies that do not show a particularly strong energy dependence. Both these implications are in accord with the facts.

The foregoing paragraphs are only a very brief summary of some of Bohr's observations at the Geneva conference. He accounts qualitatively for a number of other features of fission. His work leads one to hope

that a quantitative development of some of the ideas associated with the collective model will considerably increase our understanding of the nuclear fission process.

ACKNOWLEDGMENTS

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Decay Scheme of La^{138}

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The methods of scintillation spectroscopy have been applied to the naturally occurring radionuclide La^{138}. It is shown that La^{138} emits gamma radiation of energies 1.43±0.01 and 0.81±0.01 Mev. Coincidence studies show that the 1.43-Mev radiation is correlated in time with an observed Ba K x-ray (32±1 kev), but not with the 0.81-Mev gamma ray. A decay scheme is proposed in which La^{138} undergoes electron capture to the first excited level of Ba^{138} at 1.43±0.01 Mev with a partial half-life of $(2.1\pm0.1)\times10^{11}$ years and an L/K ratio of 1.4±0.25, and negatron decay to an excited level of Ce^{138} at 0.81 Mev with a partial half-life of $(2.4\pm0.2)\times10^{11}$ years. Consideration of beta decay systematics suggest that in each case the transition energy is low. The total half-life is estimated to be $(1.0\pm0.1)\times10^{11}$ years.

INTRODUCTION

The natural radioactivity of lanthanum has been reported[1] and confirmed.[2–4] Nuclear systematics indicate that the radioactive nuclide is the rare (isotopic abundance 0.089%)[5] isotope La^{138}, since this isotope is the central member of the naturally occurring isobaric triplet Ba^{138}–La^{138}–Ce^{138}, and is odd in both N and Z. In an earlier investigation,[2] gamma radiation was detected, of energies 1.39±0.03, 0.807±0.015, and 0.535±0.015 Mev and relative intensities 1:0.65:0.3, respectively, with a total specific gamma-ray activity of 0.6 gamma quanta per second per gram of lanthanum. In addition, the observation of barium K x-radiation (32±1 kev) of specific activity 0.4 x-rays per second per gram of lanthanum led to the proposal of a decay scheme in which La^{138} undergoes K capture to levels in Ba^{138} at 0.807 Mev and 1.39 Mev. In contrast, Bell and Cassidy[3] have since reported that the gamma-ray spectrum of La^{138} comprises two components of energies 1.06 Mev and 0.545 Mev. Mulholland and Kohman[4] have detected a low-intensity negatron component (0.07 disintegration per second per gram of lanthanum) with an end-point energy of 1.0±0.2 Mev. Their study of the x-ray spectrum yielded a specific activity of 1.0 K x-rays per second per gram. Selig[6] has studied the x-ray spectrum and reports activities of 0.23±0.07 K x-rays and fewer than 0.013 L x-rays per second per gram of lanthanum with an L/K capture ratio of less than 0.5. Recent studies[7–9] of the beta and gamma-ray spectra of Cs^{138} have shown that the first excited state of Ba^{138} is at 1.426 Mev but otherwise contradict the proposed decay scheme for La^{138}. We have re-examined the gamma-ray spectrum of lanthanum with a high-resolution scintillation spectrometer in a massive lead and mercury shield and have studied the time correlation of the observed components.

SPECTRUM ANALYSIS

For the analysis of the electromagnetic spectrum of La^{138}, a well-shielded crystal of NaI(Tl) ($1\frac{1}{4}$ in. in diameter and $1\frac{1}{2}$ in. high) mounted on a DuMont K1186 photomultiplier was used. A detailed description of the massive lead and mercury shield has been published elsewhere.[10] After amplification in a high-gain, non-overloading amplifier, the pulse-height spectrum was analyzed by means of a Harwell type-1074A five-channel descriminator. The resolution of the spectrometer for gamma radiation of 0.662 Mev was

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