Symmetry of Neutron-Induced U^{235} Fission at Individual Resonances*

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(Received December 5, 1960)

Neutrons in the resonance energy region from a nuclear explosion were resolved by time-of-flight and used to induce fissions in U^{235} attached to a revolving wheel. The symmetry of fission at individual resonances from approximately 10 to 60 ev was examined by radiochemical means. As measured by the ratio Ag^{111}/Mo^{99}, the probability of symmetric fission decreased at some resonances by a maximum of 10% compared to thermal fission of U^{235} and at other resonances increased by a maximum of 40%. With varying degrees of assurance, nine resonances are identified with an increase in symmetry; five more regions of increased symmetry are associated with resonances or a background effect. Twenty resonances are identified with a decrease in symmetry. In a sample containing 500 levels in the resonance region, there was no level with a Ag^{111} yield even one-thirtieth as great as Mo^{99}. It is thus very improbable that there are any neutron resonances in U^{235} that lead to predominantly symmetric fission.

INTRODUCTION

THE possibility of variations in the fission yield of various mass chains as a function of incident neutron energy in the low-energy fission of heavy elements is of interest for a combination of practical and scientific reasons. The question arises practically, for instance, when radiochemical analysis for a few fission products is used to infer the total number of fissions in a heavy element. If a thermal reactor is operated close to 3000°C, which is conceivable in terms of present-day technology, it would be unwarranted to assume that the yield of a given fission product is the same as at room temperature since a large number of fissions would occur in the resonance region. A more basic scientific question arises from some of the present theories[1–3] concerning the fission process which hypothesize that the compound nucleus undergoing fission may be relatively "cold" due to conversion of excitation energy to potential energy of deformation, and thus this deformed nucleus can exist only in a relatively small number of well-defined rotational and vibrational collective quantum states. In this case, low-energy neutron-induced fission might occur preponderantly through a transition state of a unique total angular momentum and parity. Depending on whether the spin of the incident neutron adds or subtracts from the spin of the nucleus, the relative symmetry of fission induced by neutrons in the resonance energy region might vary from level to level.[4] Quite apart from detailed theoretical arguments concerning the nature of the fission process, it would be of interest to determine whether each resonance has its own characteristic fission pattern or whether all resonances show a common or a very limited number of patterns. Until the question is examined experimentally, there is always the possibility of finding highly symmetric modes of fission in the resonance region.

The details of the symmetry of the fission process can be most sensitively studied by radiochemical examination of the mass distribution of the fission products. A convenient gross indicator of the mass distribution in fission is the ratio of the yield of a symmetric fission product to that of an asymmetric fission product (e.g., Ag^{111}/Mo^{99} for U^{235} fission).

The first attempt to obtain even very general information on this question, by the Los Alamos Radiochemistry Group,[5] demonstrated that the average yield of symmetric fission products in U^{235} fission induced by pile neutrons above the cadmium cutoff (energies >0.1 ev) was 10%–15% less than for fission by thermal neutrons. This experiment was repeated in another laboratory with substantially the same results.[6]

Another group[7] reported that fission induced in U^{235} by neutrons from a crystal spectrometer at 1.1, 3.1, and 9 ev was comparable in symmetry frequency with fission induced by thermalized pile neutrons. However, the experimental error was about $\pm 20\%$ and the experiment was not very sensitive to a decrease in symmetric fission yields due to a large background effect. Still the data do preclude the possibility of any large increase in symmetric yields at these resonances.

The group at the Idaho Materials Testing Reactor,[8] in a similar experiment with U^{233}, reported that the

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[1] A. Bohr, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955* (United Nations, New York, 1956), Vol. 2, p. 151.
[2] J. A. Wheeler, Conference on Neutron Physics held in Gatlinburg, Tennessee, November, 1956 [Oak Ridge National Laboratory Report ORNL-2309 (unpublished)].
[3] J. A. Wheeler, Physica **22**, 1103 (1956).
[4] L. Wilets, *Fission Proceedings of the Rehovoth Conference on Nuclear Structure*, edited by H. J. Lipkin (North-Holland Publishing Company, Amsterdam, 1958), pp. 122–33.
[5] Los Alamos Radiochemistry Group, Phys. Rev. **107**, 325 (1957).
[6] R. B. Regier, Idaho Materials Testing Reactor (private communication).
[7] R. Nasuhoglu, S. Raboy, G. R. Ringo, L. E. Glendenin, and E. P. Steinberg, Phys. Rev. **108**, 1522 (1957).
[8] R. B. Regier, W. H. Burgus, and R. L. Tromp, Phys. Rev. **113**, 1589 (1959).

Cd^{115}/Mo^{99} ratios indicate decreases in frequency of symmetric fission of $21.7\pm3.8\%$ and $41.5\pm10.3\%$ (relative to thermal fission) at the 1.8 and 2.3 ev resonances, respectively. At the 4.7 ev resonance, symmetry appeared to be the same as at thermal energies within a rather large experimental error. Symmetry decreased approximately 20% averaged over the epicadmium resonances. These investigators also reported[9] a greater than fivefold decrease in symmetry of Pu^{239} fission at the 0.297 ev resonance compared to thermal fission. The yield ratios in the epi-Sm fission of Pu^{241} were approximately the same as are observed in thermal fission.

An Argonne National Laboratory group[10] has used a double ionization chamber method of measuring the yields of different types of fission. Neutron filters were used to accentuate the proportion of resonance neutrons in a beam emerging from a pile. This group has reported an increased symmetry of fission of U^{235} when such resonance neutrons were used. These results appear to be in direct contradiction to those reported by the other groups cited above.

Progress in this field has been definitely handicapped by the lack of neutron sources providing suitable intensities in the resonance region. Radiochemical investigations, despite their excellent mass resolution, require fluxes which are orders of magnitude higher than are required in physical measurements of resonance effects. Since our calculations made it seem probable that the flux intensities at energies above a few electron volts from a small nuclear explosion were orders of magnitude greater than were available from any other known laboratory source, the Los Alamos Radiochemistry Group proposed to make use of these neutrons to study the symmetry of fission in the resonance energy region. The fluxes were calculated to be high enough to make the use of radiochemistry practical in conjunction with energy resolution by time-of-flight methods.

A nuclear fission explosion releases approximately 1.5×10^{23} neutrons per kiloton of energy over a time interval of much less than 1 μsec. The neutrons can be completely moderated in nearby hydrogenous material in times of the order of a few μsec. Ordinarily, this moderator will be heated by prompt gamma radiation, x rays, and fast neutrons to a very high temperature and the neutrons will tend to moderate to a thermal distribution in the resonance region. This temperature can, in principle, be controlled by varying the amount of shielding between the moderator and the explosion. The amount of moderation and average time delay for moderation can be controlled by varying the thickness of moderator. The moderated neutrons are released when the blast wave from the explosion passes through the material.

Because of the uniquely large source available from a one kiloton device, the flight path can be hundreds of meters, with correspondingly high resolution of energy by time-of-flight. For physical measurements, where intensity can be sacrificed for energy resolution, resolutions of the order of 10^{-3} μsec/m should be practically achievable over a flight path of 500 m at energies above 10 ev. For radiochemical work, intensity is an important consideration and requires some compromise of resolution.

In the experiment to be described in this paper a wheel faced with U^{235} was rotated in front of a slit defining the beam of neutrons from the explosion. The fission products from fission produced by neutrons at various energies were therefore to be found in different parts of the wheel and could be examined radiochemically after recovery of the apparatus.

EXPERIMENTAL PROCEDURES

The experiment was first attempted in May, 1958, at Eniwetok. Although the results obtained were comparatively poor due to insufficient neutron shielding around the target, the flux of resonance neutrons reaching the target was in the range predicted. A similar experiment was performed in October, 1958, in Nevada. The nuclear device utilized had an energy yield corresponding to 84 tons of TNT. The geometry

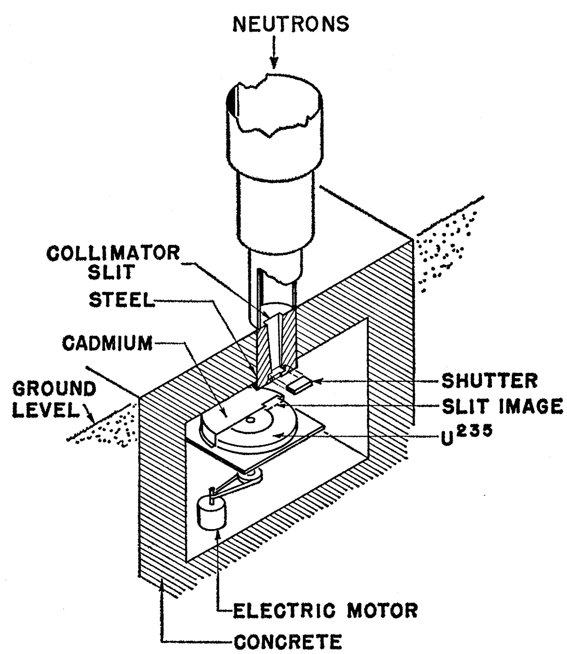

FIG. 1. Schematic arrangement of apparatus for time-of-flight separation of resonance neutrons from a nuclear explosion as used in Nevada in October, 1958. The essential components and their relative orientation are indicated, but the dimensions are not to scale. The position of the nuclear device and surrounding moderator was above the apparatus (not shown). The tapered pipe was evacuated with the ends closed by thin aluminum.

[9] R. B. Regier, W. H. Burgus, R. L. Tromp, and B. H. Sorensen, Phys. Rev. **119**, 2017 (1960).

[10] L. W. Roeland, L. M. Bollinger, and G. E. Thomas, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 440.

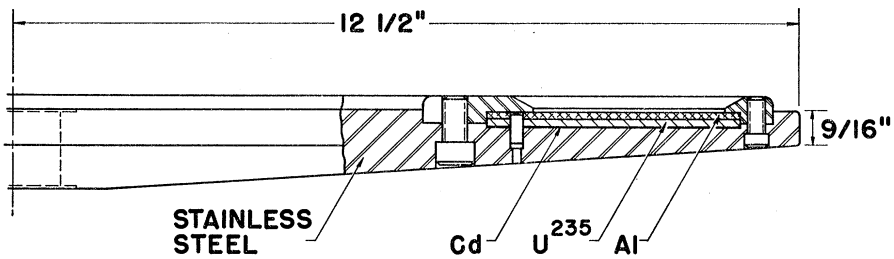

FIG. 2. Cross section of wheel assembly for time-of-flight experiment. The U^{235} layer was actually composed of six foils, as described in text.

and equipment are shown schematically in Fig. 1. The distance from the explosion source to the target was 3.13×10^3 cm. A doubly tapered collimating slit 10 cm long and 0.10 cm wide through a 5-ft thick steel mask defined a beam of neutrons which impinged on 93% U^{235} metal fastened to the outer rim of a 2-ft diam wheel. The outer radius of the metal was 29.21 cm; the portion of the metal exposed to neutrons extended from a radius of 21.0 to 28.5 cm. The wheel was revolving with a precisely measured speed of 67.68 rps when the nuclear device exploded. It is shown in detail in Fig. 2. A very heavily shielded bunker protected the target from extraneous neutrons and blast. Most of the neutron flight path was contained in a tapered evacuated pipe which reached a 3-ft diam at the source end. Further collimation of neutrons was achieved by surrounding the pipe midway between source and target with boron-loaded plastic. The bunker was lined with boral to absorb epicadmium neutrons. The wheel was enclosed in a cadmium metal box with a slit in it to permit the beam to reach the wheel. After the explosion, the exposure of the wheel to neutrons was terminated by a cadmium-covered steel shutter which was moved by explosive squibs across the collimating slit. This shutter was closed after the wheel had turned a half-revolution following the instant of the explosion. The shutter stopped neutrons below the cadmium cutoff from reaching the wheel and also intercepted debris that might come down the pipe.

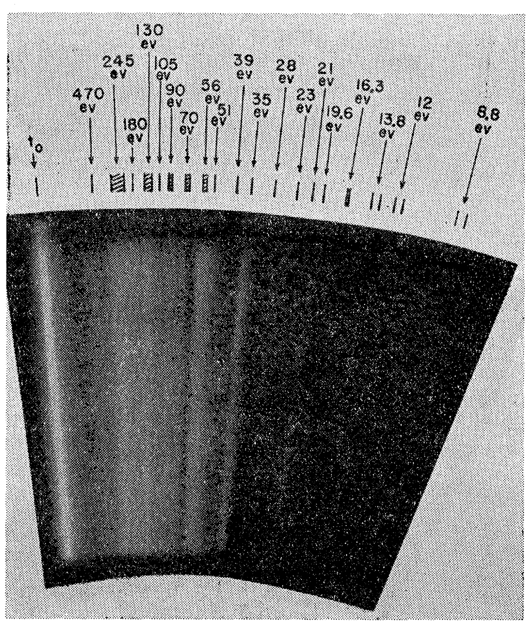

FIG. 3. Contact radioautograph of topmost 2-mil U^{235} foil on wheel in 1958 experiment. The light features in the figure correspond to blackening of the photographic plate. The motion of the wheel was from left to right. The bright line at the extreme left is due to fissions from prompt neutrons, (t_0). The structure to the right of this shows the variation in fission activation of the wheel due to resonances in the fission cross section of U^{235}. The positions of known maxima in this cross section are indicated by the figures above the radioautograph.

The beam width actually defined by the collimating slit on the wheel was somewhat greater than the 0.10 cm width of the slit. The wheel was 11.4 cm from the defining slit. At this distance the maximum width of the beam defined by the 0.10 cm slit for a diffuse source uniformly filling the field of view at the source end is 0.24 cm. The actual source did not uniformly fill the field of view. The sharpest structure observed in the resonance region of the radioautograph was approximately 0.15 cm wide.

The U^{235} metal portion of the wheel consisted of a sandwiched set of six foils with a total thickness of 0.248 cm. The top foil was 0.00551 cm thick (nominally 2 mils), the second 0.0258 cm (10 mils), and the next four foils 0.0542 cm thick (20 mils), respectively. The density of this metal was 18.82 g/cm^3 corresponding to 4.82×10^{22} atoms of uranium per cm^3. The top uranium foil was covered with 0.125-in. aluminum sheet and the whole sandwich was backed with 0.016-in. cadmium metal, all clamped to an approximately $\frac{1}{2}$-in. thick stainless steel wheel.

The neutrons were moderated in a hydrogenous material close to the fissioning core and escaped over a time interval of the order of 1 μsec. The time interval for moderation corresponds to a maximum uncertainty of about 0.01 cm in the position of neutrons of a given energy on the wheel. Unmoderated neutrons with fission spectrum energies defined a line on the wheel at a position S_0, at an almost instantaneous time t_0. Neutrons in the resonance region required times of the order of a millisecond to arrive at the wheel. The distance from S_0 on the perimeter of the wheel as a function of neutron energy is given by

$$S=V_{\mathrm{wh}}D/V_nE_n^{\frac{1}{2}},$$

where V_{wh}=velocity of wheel at rim, D=distance be-

TABLE I. Representative sample schedule and experimental results (30–45 ev).

Sample no.	Distance on wheel (cm) S_1	S_2	$\bar{S}$	$\bar{E}_n$ (ev)	Mass (mg)	$A_{Mo^{99}}$/mg U^{235} (in counts/min)	$A_{Ag^{111}}/A_{Mo^{99}}$ (in units of 10^{-3})	$A_{Ba^{140}}/A_{Mo^{99}}$
62	4.11	4.20	4.15	45.6	384.6	147.5	1.11	0.429
63	4.20	4.29	4.24	43.7	362.0	146.8	1.06	0.408
64	4.29	4.38	4.33	41.9	358.4	139.5	1.08	0.400
65	4.38	4.47	4.42	40.2	367.7	137.2	1.15	0.448
66	4.47	4.57	4.52	38.6	383.1	132.5	1.36	0.420
67	4.57	4.66	4.61	37.0	353.0	109.5	1.31	0.412
68	4.66	4.78	4.71	35.4	493.7	164.5	1.36	0.415
69	4.78	4.89	4.83	33.8	433.7	173.0	1.14	0.411
70	4.89	4.98	4.93	32.4	362.3	132.5	0.99	0.447
71	4.98	5.07	5.02	31.2	375.5	111.2	0.91	0.436
72	5.07	5.16	5.11	30.1	358.4	94.8	1.06	0.427

tween source and target, V_n=velocity of 1-ev neutron, and E_n=energy of neutron in ev.

Under the conditions of this experiment the combined factors give

$$S(\text{cm})=28.08/E_n^{\frac{1}{2}},$$

and $dS/dE=-14.04/E^{\frac{3}{2}}$ at the rim or $-11.7/E^{\frac{3}{2}}$ at the average cut radius.

Thus, for example, a 10-ev neutron struck the wheel rim at a distance of 8.88 cm from S_0; the linear separation of neutrons at the rim of the wheel at 10 ev was 0.44 cm/ev. The 1-mm slit gave a theoretical resolution of 0.27 ev at the middle of a cut at the 10-ev position and 17.3 ev at the 160-ev position.

After recovery, delayed for almost a week by collapse of the bunker roof, the top foil was removed and used to make an autoradiograph. This is reproduced in Fig. 3. The structure in this picture is due to variations in fission product density corresponding to resonances in the epithermal energy region. The time t_0 is defined by a sharp line due to fissions induced by fast neutrons arriving within a few μsec after explosion time.

With the aid of this picture, the regions of interest on the next two foils were cut into radial slices, for the most part 0.1 cm wide at the rim. Time and available analytical effort limited the number of analyses, particularly on the 20-mil foil. These samples were first counted for gross fission product activity by a NaI gamma scintillation counter and then dissolved and analyzed for several fission products by standard radiochemical methods.[11] The schedule of cuts is illustrated in Table I. The first column of Table I gives the sample number designation of the radial slices. The next three columns establish the position of the slice relative to the prompt neutron line on the wheel. The distances were measured along the perimeter of the uranium facing of the wheel. The fifth column, labeled $\bar{E}_n$, gives the average energy of the neutrons impinging on the slice. The sixth column indicates the weight of uranium represented by the slice. Space limitations do not permit the complete tabulation of these data for approximately 175 samples but they are summarized in Fig. 4.

It was not attempted in this experiment to determine the absolute fission yield of any fission product. Although it was assumed that the high-yield fission products are produced in nearly constant yield throughout the resonance region, an effort was made to partially check the assumption by measuring both Ba^{140} and Mo^{99}. It was expected that if the ratio Ba^{140}/Mo^{99} corresponded to that observed for thermal neutron fission, it would be reasonable to conclude that the individual fission yields had probably not changed.

The yield of Ag^{111} was measured to determine the symmetry of fission. Changes in the value of the ratio of Ag^{111} to that of Mo^{99} relative to that observed in the thermal neutron fission of U^{235} are interpreted as changes in fission symmetry relative to the known characteristics in thermal neutron fission. An effort was made to determine changes in the light-mass wing of the yield distribution by analysis for As^{77} but the very poor statistics make the data inconclusive. Because of the delay in recovery of the wheel, several interesting short-lived fission product activities could not be used. A few results on Cd^{115} in general corroborated the results from Ag^{111}.

Resolution was lost in the mechanical cutting of the wheel due partly to the improbability of including all of a particular resonance in a single cut and excluding the neighboring valley region, and partly to the difficulty of cutting each metal strip exactly on a radius.

EXPERIMENTAL RESULTS

Results for Mo^{99}, Ag^{111}, and Ba^{140} are illustrated in Table I. The seventh column of Table I gives the Mo^{99} activity in each slice of the wheel in counts per minute per milligram of U^{235} at the time of the explosion under standard counting conditions and corrected for chemical recovery. The eighth column gives the ratio of Ag^{111} to Mo^{99} radioactivity in the slice under the counting conditions used. This ratio, under the same conditions, for the thermal neutron fission of U^{235}, is 1.0×10^{-3}. Finally, the last column gives the ratio of Ba^{140} to Mo^{99}

[11] J. Kleinberg *et al.*, Los Alamos Scientific Laboratory Report, LA-1721, (unpublished) 2nd ed. (1958).

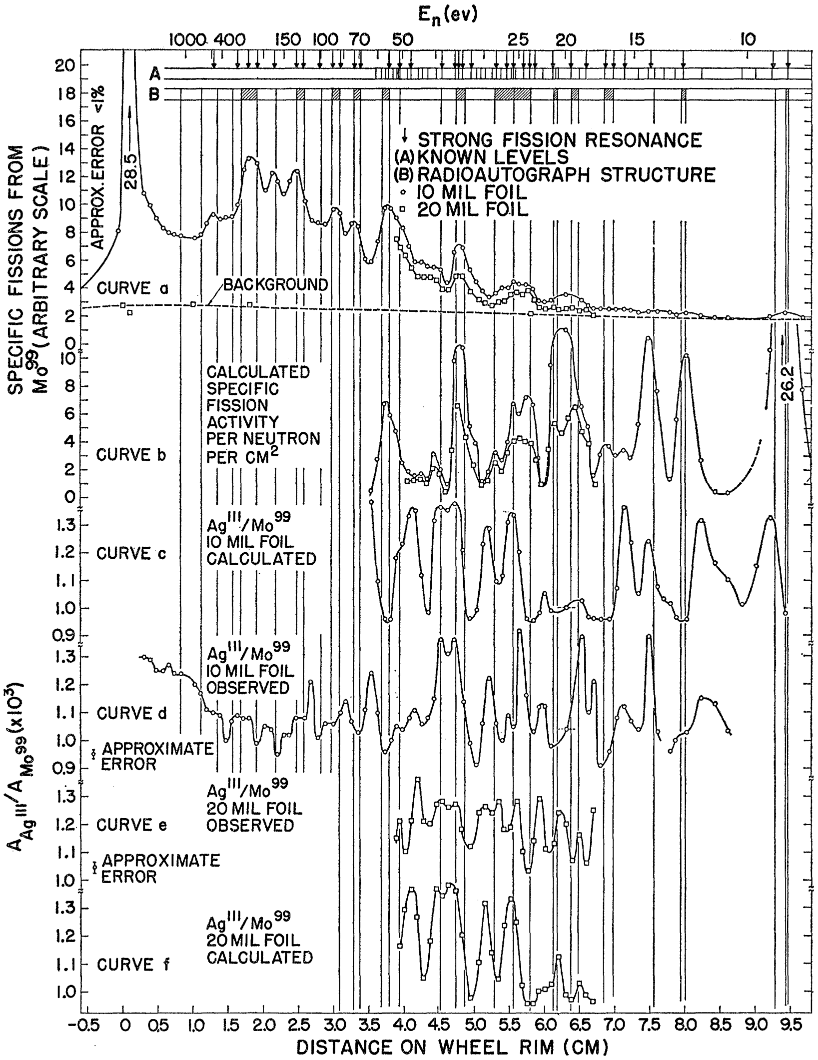

FIG. 4. Experimental results on fission density and Ag^{111}/Mo^{99} activity ratios from time-of-flight experiment. The ordinates are the distance on the wheel rim on the bottom and the corresponding energy of the neutrons hitting the wheel at this point (top ordinate). The arrows just below the top ordinate indicate the position of strong maxima in the fission cross section of U^{235}. The section A shows the position of all known maxima in the fission cross section of U^{235}. Section B shows the observed regions of high fission density on the radioautograph. These are carried down through the rest of the figure by vertical lines to facilitate comparison of structure in the curves with observed levels. Curve a is a plot of the fission density (as measured by the Mo^{99} specific activity) in the 10-mil U foil as a function of perimeter distance in the wheel. The units are arbitrary. The lower curve, in the region 4–7 cm represents the data on the 20-mil piece. Also indicated, by the approximately horizontal dashed line, is the background level of fissions outside the area illuminated by the slit. Curves d and e are observed Ag^{111}/Mo^{99} activity ratios in the 10- and 20-mil U^{235} foils, respectively, as a function of distance on the wheel. Curves b, c, and f are the calculated fission densities and the Ag^{111}/Mo^{99} activity ratios in the 10- and 20-mil U^{235} foils.

radioactivity in the slice. The corresponding value in thermal neutron fission is 0.42.

The experimentally measured values for Mo^{99} specific activity as a function of distance on wheel (i.e., neutron energy) are presented graphically in curve a of Fig. 4. Actual counting rates varied from a few hundred counts per minute of Mo^{99} to 10 counts/min of Ag^{111} in a given sample. Although the specific activities are for cuts of finite width, they are plotted as a continuous distribution since the cuts are generally so narrow that a histogram would approximate a smoothed curve.

Background fissions were produced by neutrons which penetrated the shielding and by fission neutrons produced in the wheel. These fissions were measured by analysis of samples taken from the wheel rim adjacent to the slit image. In addition, analyses were made of U^{235} from a portion of the wheel which was never exposed to the open collimating slit. Unfortunately, the analyses for Ag^{111} were not useful due to insufficient activity for good statistics. However, the Mo^{99} specific activity indicates the contribution of general background to observed signal and is indicated in curve a of Fig. 4 by a dotted line.

It is seen that these background fissions were rather uniform over the wheel. They were only a small part of the signal recorded in the region above 30 ev but were the main effect below about 18 ev.

In the topmost part of Fig. 4, above the graph of the experimental results, there are indicated the positions of known strong fission resonances (shown by arrows on the figure). These energies are taken primarily from the Brookhaven compilations.[12] Also at the level *A* of the figure there are given the positions of all known resonance levels in the fission cross section of

[12] *Neutron Cross Sections,* compiled by D. J. Hughes and J. A. Harvey, Brookhaven National Laboratory Report BNL-325, (U. S. Government Printing Office, Washington, D. C., 1957 and 1958), Suppl. No. 1 and 2nd ed.

U^{235} as tabulated primarily by Havens *et al.*[13,14] Finally, at the level B at the top of Fig. 4, there are shown the regions of blackening on the radioautograph.

It should be noted that the radioautograph shows more features of the wheel activation than the chemical analyses, partly for reasons that have already been discussed as causing loss of resolution in the cutting of the wheel. In addition, the radioautograph can be examined at the rim, where the resolution is highest; it was made from the uppermost foil, before scattering and absorption occurred; and the eye is extremely sensitive to small relative differences of intensity on a photographic plate.

DISCUSSION

It seems quite clear that fissions induced in U^{235} by neutrons of various energies in the resonance region have been isolated. Not only are the variations in fission density as a function of distance on the wheel strongly suggestive of cross section variations but the plot of the radiochemically measured fission density structure (curve a, Fig. 4) corresponds, over the energy region from 8.8 ev to $\sim$500 ev, to most of the known macroscopic features of the U^{235} fission cross section (arrows at the top of Fig. 4 and curve b, Fig. 4). In particular, the last discernible feature on the radioautograph, which is barely visible on Fig. 3, is the well-isolated line at 9.45 cm from S_0 which corresponds precisely to the expected position of the 8.8-ev resonance, confirming the validity of the wheel equation. Moreover, in the regions where analysis of the 20-mil foil were performed (20 to 50 ev), the variation observed in fission density as a function of distance on the 10-mil foil were confirmed (curve identified with square data points, curve a Fig. 4).

A plot of the measured Ag^{111} specific activity would also show structure but not in exactly constant ratio to Mo^{99}. As a result, plots of the Ag^{111}/Mo^{99} ratio (curves d and e, Fig. 4) show structure as a function of neutron energy. This ratio varies from 0.9×10^{-3} to 1.4×10^{-3} in the resonance region.

The larger variations in the Ag^{111}/Mo^{99} ratios correspond sufficiently in the region where two foils were analyzed so that they may be accepted for the most part as physically real and not merely the result of statistical fluctuation and analytical error. As mentioned above, experimental error in the determination of these ratios may be estimated from previous experience as of the order of $\pm5\%$, somewhat less for the highest counting samples and somewhat more for the lowest counting samples. Although the data on the third foil (20 mil) show significantly higher ratios and less peak-to-valley variation than are observed in the second foil (10 mil), the maxima and minima in the two foils are definitely related.

Whereas there is, in general, good correlation between positions of high-fission density and calculated positions of high-fission cross section, the correlation of extrema in the Ag^{111}/Mo^{99} ratios with positions of high-fission cross section is somewhat less marked. For example, of the five highest Ag^{111}/Mo^{99} ratios, four occur (at approximately 14, 25, 35, and 39 ev) where there are known maxima in the fission cross sections. On the other hand there are other peaks and minima which are not related to known cross section maxima.

It is clear from examination of the top part of Fig. 4 that the most intensity in this experiment was in the region above 20 ev where the resolution did not clearly separate individual resonances with their average spacing of 0.7 ev. On the other hand both the gross fission activity (as measured by Mo^{99}) and the Ag^{111}/Mo^{99} ratios show structure in this region. The simplest hypothesis to adopt, in an effort to explain these variations, is that each resonance is characterized not only by the usual parameters of fission width, neutron width, etc., but also by one of two fission product yield distributions. In order to establish whether such a hypothesis would explain the observed variations of the Ag^{111}/Mo^{99} ratios, a series of calculations were made using an IBM 704 EDPM. In the calculation reasonable resonance parameters for levels with energy less than 60 ev were combined with instrument resolution parameters to calculate the expected fission density in the wheel in a flux having uniform density per unit energy.

The calculations have been performed in the following way:

The cross sections associated with each resonance were calculated from the Breit-Wigner single-level formula for neutron absorption and for fission. The values used for E_0 at each resonance and the resonance parameters are an eclectic set coming, in large part, from the authors interpretation of some data reported by Havens *et al.*[13] and from BNL-325. The values used do not agree exactly with any other published set. They are not represented as "best" values but only as consistent with the recently published Columbia values[14] and with the indications of our own data. The principal differences have to do with the assignment, for purposes of this calculation, of energies and widths to levels which are elsewhere reported as not resolved. Noticeable differences between our data and those of Havens exist at 14.1, 18.7, and 22.1 ev which were assigned resonances in this paper. Otherwise, Ag^{111}/Mo^{99} curves computed with input data taken entirely from Havens *et al.*[14] do not differ significantly from those presented in this report. The input data are presented in Table II.

In this table the second and third columns give the assumed values of the fission width (Γ_F) and the neutron width (Γ_N) in millivolts for the resonances whose neutron energy characteristics in electron volts are listed in the first column. The gamma width ($\Gamma\gamma$) was

[13] W. W. Havens *et al.* Columbia University Progress Report, January–March, 1958 (unpublished).

[14] W. W. Havens, E. Melkonian, L. J. Rainwater, and J. L. Rosen, Phys. Rev. **116**, 1538 (1959).

TABLE II. Ingoing data for IBM 704 calculation and final symmetry assignment.

Resonance energy E_0	Γ_F (ev in units of 10^{-3})	Γ_n (ev in units of 10^{-3})	S_0 (cm)	Assigned Ag^{111}/Mo^{99} (in units of 10^{-3})
0.282	98	0.0037	52.9	1.175
1.14	107	0.0147	26.3	1.175
2.04	120	0.0066	19.7	1.175
2.82	6	0.0025	16.7	1.175
3.14	115	0.0284	15.8	1.175
3.60	45	0.0532	14.8	1.175
4.85	4	0.0550	12.8	1.175
5.45	6	0.0210	12.0	1.175
5.83	6	0.0160	11.6	1.175
6.10	6	0.0272	11.4	1.175
6.39	6.47	0.296	11.1	1.175
7.10	16.4	0.109	10.5	0.95
8.78	30.3	1.28	9.48	0.95
9.26	110	0.122	9.23	1.40
9.73	6	0.0405	9.00	0.95
10.15	6	0.0638	8.81	0.95
11.65	2.6	0.683	8.23	1.40
12.40	12.3	1.374	7.97	0.95
12.80	6	0.0507	7.85	0.95
13.35	6	0.0987	7.69	0.95
13.80	98	0.149	7.56	0.95
14.10	166	0.199	7.48	1.40
14.65	42.5	0.153	7.34	0.95
15.5	17.5	0.232	7.13	1.40
16.2	9.4	0.338	6.98	0.95
16.8	22.9	0.275	6.85	0.95
18.2	48.2	0.341	6.58	0.95
18.7	10.1	0.199	6.49	1.40
19.4	41.0	2.91	6.38	0.95
20.6	21.9	0.363	6.19	1.40
20.9	5	0.500	6.14	0.95
21.1	32.3	1.10	6.11	0.95
22.1	6	0.0893	5.97	1.40
23.0	16.4	0.671	5.86	0.95
23.6	18.0	1.60	5.78	0.95
24.4	38.5	0.494	5.68	0.95
25.3	19.8	0.382	5.58	1.40
25.6	31.3	0.608	5.55	1.40
25.9	8.2	0.407	5.52	0.95
26.5	15.0	0.510	5.45	1.40
27.3	6	0.110	5.37	0.95
27.9	25.3	0.740	5.32	0.95
28.6	10.7	0.374	5.25	1.40
29.7	6	0.380	5.15	1.40
30.4	7.7	0.386	5.09	1.40
31.1	11.0	0.669	5.04	0.95
32.2	29.2	1.70	4.95	0.95
33.7	24.5	2.09	4.84	0.95
34.6	23.0	3.56	4.77	1.40
35.3	34.6	5.94	4.73	1.40
38.4	6	0.161	4.53	0.95
39.7	26.2	2.52	4.46	1.40
42.0	11.7	2.27	4.33	0.95
43.7	12.4	0.992	4.25	0.95
44.8	15.1	1.81	4.20	1.40
47.1	23.2	1.24	4.09	1.40
48.6	5.5	0.523	4.03	0.95
50.8	4	3	3.94	0.95
51.6	14.0	6	3.91	1.40
52.3	8	3	3.88	0.95
53.5	3	2	3.84	0.95
55.4	84	2.90	3.77	0.95
56.4	27.6	8.63	3.74	0.95
58.3	28.7	3.82	3.68	0.95
61.0	124	1.17	3.60	1.40

taken constant at 0.033 ev. The fourth column gives the distance of the resonance on the wheel from the prompt neutron line.

The energy resolution was assumed to be limited by the width of the collimating slit and was calculated for a uniform distribution of neutrons in a slit 0.16 cm wide (60% greater than the actual slit). Activations were calculated over this distribution in 0.008-cm intervals at energy intervals corresponding to every 0.01 cm on the wheel. Summations were made for actual cut intervals over the energy region from approximately 8 to 65 ev. At higher energies resonance parameters for single levels are no longer available. The neutron flux was treated as constant with energy. No correction was made for background attributable to the various sources discussed elsewhere in this report.

The results of the calculation for fission density versus wheel position and neutron energy are shown in curve b of Fig. 4. Since the calculation presents a fission probability per incident neutron, it does not reproduce the flux variation on the wheel. It is seen, however, that the main features of the excitation function and of the observed wheel activation in the region studied are reproduced in the calculation. The observed Mo^{99} activity at the position corresponding to 35 ev indicates a time-integrated neutron flux of about 10^{10} n/cm^2 ev incident at this point.

The assumption was then made that at each level the ratio of Ag^{111} activity to Mo^{99} activity was either 0.95 or 1.4 ($\times 10^{-3}$) except that an average value was assumed for resonances from 0.3 to 6.4 ev. The choice of one or another value for the characteristic Ag^{111}/Mo^{99} ratio at each level was varied until the structure in the computed Ag^{111}/Mo^{99} curve qualitatively fitted the observed structure. The final "best" assignment of levels is listed in column five of Table II. These assignments are not unique in all cases since, for many very weak and questionable levels, no noticeable difference is made in the fit if the assignment is changed. The computed curves are shown in curves c and f of Fig. 4.

The agreement between the calculated and observed Ag^{111}/Mo^{99} ratio is fair. There is satisfactory reproduction of even fine details in the 10-mil foil data between 35 and 20 ev, and also below 15 ev. In the other regions the representation is only qualitatively satisfactory. The agreement between calculated and observed ratios in the 20-mil piece is similar, except that here the damping effect of increased background in the experimental data is clearly visible.

The observed data could obviously be fitted more closely by assuming more than two Ag^{111}/Mo^{99} values or by making this ratio arbitrarily variable. However, because very small differences in the actual position of the cuts change the resolution from resonance to resonance in an unknown way and because the background corrections are not well known, the assumption of more than two values for the Ag^{111}/Mo^{99} ratios does not appear to be justified. A background correction, if applied, would lead to assignment of more extreme values of the Ag^{111}/Mo^{99} ratios than were assumed in the calculation.

Attention has already been directed to the existence of a general neutron background which produced fissions outside the region illuminated by the collimating slit. The level of this background could be determined and is indicated by the dotted line below curve a of Fig. 4. There is some evidence that the actual background in the illuminated region may have been higher than this although the precise level cannot be estimated. This is suggested by the fact that the intensity variation in this region is not as great as predicted from calculations based on the cross sections (compare curve b of Fig. 4 with curve a). In addition, the Ag^{111}/Mo^{99} ratios in the 20-mil foil, averaged over 1 cm distances, run 5 to 8% higher than in the corresponding region of the 10-mil foil. Since the specific fission activity in the 20-mil foil was systematically lower than in the 10-mil foil, this indicates a background with a higher Ag^{111}/Mo^{99} ratio than that characteristic in general of resonance fission. Unfortunately, this higher Ag^{111}/Mo^{99} ratio in the 20-mil foil could also be due to the greater importance of laboratory contamination to the Ag^{111} analyses of this foil. The analyses of this foil were performed later and so were more subject to such contamination. The presence of some laboratory contamination at this stage was demonstrated although its constancy and exact time of appearance are in doubt.

Thus, although the existence of an enhanced background seems likely, its amount cannot be established. The two most probable sources of this background are: (1) secondary fissions induced in the wheel by neutrons born in the wheel, and (2) lack of sufficient shielding allowing noncollimated neutrons to hit the wheel.

In performing the calculation interpreting the results, we have ignored the possible effects of this background. In doing so we have left out of consideration a possible point of view that very broad resonances might contribute a background cross section on top of which the sharp resonances occur. There is no strong evidence in our data to demonstrate that another kind of fission is important at gaps between sharp resonances, but neither can this hypothesis be excluded. On the other hand, consideration of these background effects cannot affect our general conclusion that variation in the Ag^{111}/Mo^{99} ratio occurs in the resonance region and that this can be moderately satisfactorily explained, to within the present state of the data, by the assignment of two different Ag^{111}/Mo^{99} ratios to individual resonances.

The final assignments of the 54 levels between 7 and 61 ev lead to almost twice as many levels having the lower Ag^{111}/Mo^{99} value than have the higher Ag^{111}/Mo^{99} ratio. The value of 0.95×10^{-3} adopted for one of these ratios is probably an upper limit due to the effect of background (see above) and so the preponderance of levels with enhanced asymmetry of fission is consistent with published work on the symmetry of fission in the resonance region.[5]

Although many of the specific assignments made in the calculation are somewhat arbitrary, there are regions where the assignments cannot be changed without drastic disagreement with the experimental data. The resonances which we believe have been identified with considerable assurance as possessing increased symmetry, relative to thermal fission, occur at approximately 25, 34.4, 35.3, 39.5, and 44.7 ev. With somewhat less assurance, we have identified other resonances with increased symmetry of fission at 11.6 ev, approximately 14.1, 15.4, and 51.6 ev. Increased symmetry of fission, related either to resonances or to a background effect, is observed at 18.7, 20.7, 22.1, 29, and 63.3 ev. The following resonances have been identified with decreased symmetry: 18.2, 19.4, 21.1, 23.6, approximately 26, 32.1, 33.6, 38.4, 41.9, 43.5, and 48.6 ev. In addition, with less assurance, resonances which are identified with decreased symmetry occur at 12.4, 13.8, 14.6, 16.8, 22.9, 24.4, 27.8, 30.9, and 56.4 ev.

Thus, in the region from 10 to 63 ev, we have identified five resonances which, with reasonable certainty, are associated with an increase in fission symmetry, another four resonances which are probably associated with increased symmetry, and five more regions of increased symmetry which are related to resonances or to a background effect. This background effect, if it exists, does not uniformly indicate increased symmetry whenever the specific activity is low. In addition, eleven resonances are identified which, with reasonable certainty, are associated with a decrease in symmetry, and nine more resonances are identified which are probably associated with a decrease in symmetry.

At energies above 63 ev there are no published detailed data on the parameters of individual resonances. Our data indicate regions of decreased symmetry at 71 ± 2 ev, 103 ± 3 ev, 163 ± 6 ev, 216 ± 12 ev, and 369 ± 20 ev. All of these features coincide with more or less well-defined structure in the fission excitation function for U^{235} as presented in BNL-325, 2nd ed. In addition there are regions of increased symmetry at 79 ± 2 ev and 110 ± 4 ev which appear to coincide with low-specific fission activity although neighboring samples of equally low specific activity do not show equally high Ag^{111}/Mo^{99} values.

The Ba^{140}/Mo^{99} values show no systematic or repeated structure which would indicate that the fission yield of one of them changed with respect to the other as a function of neutron energy. A rather large spread of values for this ratio is due mostly to errors in the Ba^{140} analysis arising from contamination by uranium daughter products in the heavy metal samples taken for analysis.

The upper limit for symmetry of fission at any single level in the energy region from 10 to 400 ev can be estimated by assuming that, in the region of 400 ev, where a cut 0.096 cm wide was taken, about 100 levels could have been included. For one of these levels to increase the average yield of Ag^{111} in the piece by 10% would require that it have a fission yield of $\sim$0.1% if the average yield of the other levels were the same as at thermal, or $\sim$0.2% if the average yield of the other levels were 10% lower than thermal. Since this is the upper limit to the change observed in this region, it may be said that in a sample of about 500 levels, there was no level which produced fission so highly symmetric that the Ag^{111} yield was increased twenty-fold over that found in thermal-neutron-induced fission. Since even this limit would lead to a Ag^{111} yield of only 0.2%, we can confidently conclude that none of the first 500 resonances of U^{235} gives rise to symmetrical fission.

ACKNOWLEDGMENTS

We wish to acknowledge the assistance of W. B. Goad, who performed calculations on conditions required for optimizing neutron fluxes in the resonance energy region from nuclear bomb explosions; of A. L. Bond and E. E. Shaw, who contributed their engineering and field supervisory services; of F. J. Waddell, who made the radioautograph reproduced in this report; and of J. T. Mann and many others who supported the machine computation effort. The services of a large number of other people at the Los Alamos Scientific Laboratory, particularly members of the Chemistry and Metallurgy Division who fabricated and cut the wheel after bombardment, were essential to the success of the experiment and are gratefully acknowledged.

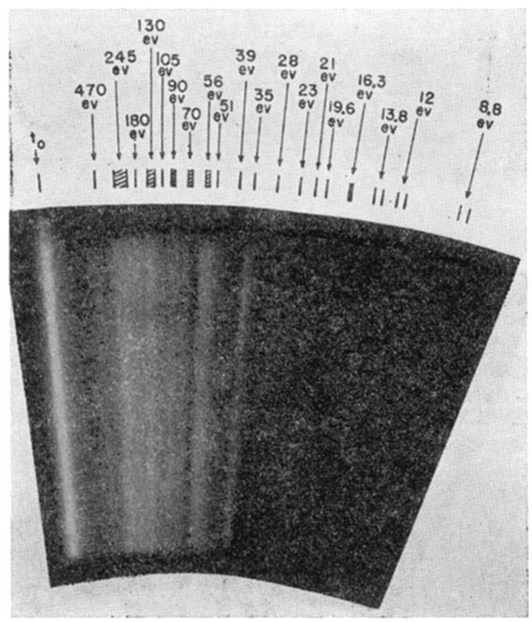

FIG. 3. Contact radioautograph of topmost 2-mil U^{235} foil on wheel in 1958 experiment. The light features in the figure correspond to blackening of the photographic plate. The motion of the wheel was from left to right. The bright line at the extreme left is due to fissions from prompt neutrons, (t_0). The structure to the right of this shows the variation in fission activation of the wheel due to resonances in the fission cross section of U^{235}. The positions of known maxima in this cross section are indicated by the figures above the radioautograph.