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

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The symmetry of neutron-induced fission in ^{235}U has been measured at a large number of epithermal levels with a time-of-flight energy-resolved beam from an underground nuclear explosion. The energy resolution was improved over previous similar "wheel" experiments. The symmetry of fission, as measured radiochemically by the ratio of ^{115}Cd to ^{99}Mo, shows a bimodal distribution. If the results are averaged separately for the two apparent groups, the first group (I) has an average ^{115}Cd/^{99}Mo ratio which is 0.593 times the thermal value; the second-group (II) average is 1.11 times the thermal value. The average fission width in I is one-half the average fission width in II. The numbers of levels assigned to I and II are 24 and 14, respectively. The energies of levels assigned to I are 19.30, 21.07, 22.94, 23.63, 26.44, 27.81, 30.91, 32.10, 33.55, 34.85, 40.51, 41.91, 43.41, 44.04, 45.79, 46.93, 48.06, 48.82, 51.37, 56.58, 57.83, 58.15, 60.25, and 61.10 eV; assignments to II are at 24.23, 25.65, 28.35, 34.39, 35.21, 35.75, 36.64, 38.42, 39.44, 44.75, 49.51, 52.27, 53.56, and 58.68 eV. From analogy with conclusions based on a similar set of observations in the fission of ^{239}Pu and on arguments derived from fission-channel theory, it is hypothesized that $J=4$ in I and 3 in II.

I. INTRODUCTION

A number of experiments have been described in recent years which document variations in the yields of symmetric or near-symmetric fission products at resonances in ^{235}U and ^{239}Pu neutron-induced fission. In the case of ^{239}Pu fission it has been demonstrated in a statistically significant sample of s-wave neutron resonances ($J^\pi = 0^+$ or 1^+) that the 0^+ levels have a characteristic ^{115}Cd yield which is a factor of 4 higher than the yield at 1^+ levels.[1] The fission widths of the $J=0$ levels are larger than the $J=1$ levels by a factor of 10. The populations of the two groups are in reasonable agreement with the expected $(2J+1)$ distributions. Previous efforts to obtain equally detailed data in ^{235}U fission[2] and ^{233}U fission by the "wheel" technique have not been entirely successful due in large part to the high-level densities in the epithermal excitation functions of these nuclides and the consequent difficulty in characterizing fission yields in a sufficiently large and well-resolved sample of levels. In a recent "wheel" experiment with a ^{235}U target, the energy resolution was sufficiently improved in the region of 20–60 eV to allow characterization of a sample of 38 reasonably well-resolved levels by their relative symmetry of fission.

II. EXPERIMENTAL PROCEDURES AND RESULTS

The details of the experimental technique have been described previously.[1] In this experiment the flight path was 240.11 m long; the collimating slit was 0.2 cm wide at the rim and 7.1 cm long, the uranium metal target (~93.3% ^{235}U) was 0.381 mm thick; the wheel speed was 1.496×10^4 cm/sec at the rim; and the exposure time for a given atom on the target was 13.37 μsec, corresponding to an energy resolution of 0.0557 μsec/m (the pulse width in time of the emergent neutrons at the moderated source is neglected). Neutrons were moderated at the source to a broad epithermal distribution peaking at $\sim5\times10^{10}$/cm^2 in 13.37 μsec in the 40- to 70-eV energy region (Fig. 1).[3] The neutron flux on the wheel was terminated before the wheel had turned a full revolution when ground shock initiated by the nuclear explosion collapsed the line-of-sight pipe.

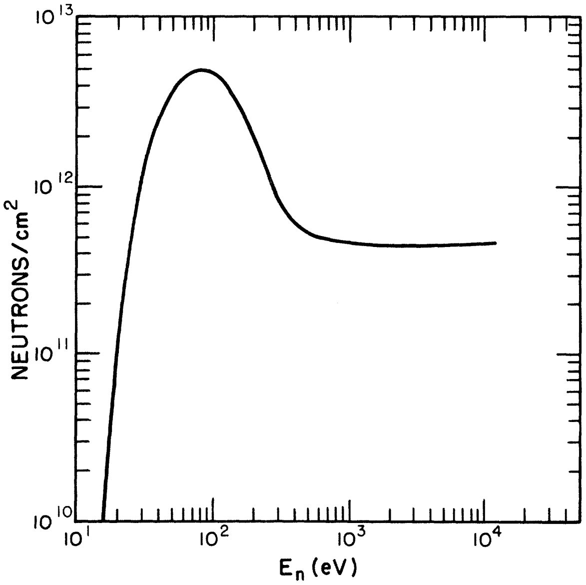

FIG. 1. $E_n(dN/dE)$ or number of neutrons per cm^2 plotted as a function of E_n (neutron energy) for the neutron beam at the target.

TABLE I. Sampled energy intervals, specific fissions in sample, $R_{\mathrm{Cd/Mo}}$, and related resonance parameters.

Energy interval					
E_1 (eV)	E_2 (eV)	Corrected fissions/g (units of 10^9)	Corrected $R_{\mathrm{Cd/Mo}}$ (±% standard deviation)	Major resonance energy[a] (eV)	ν (meV)[b]
19.33	19.60	0.112	0.402 (26.3)	19.30	70
19.86	20.07	0.036	0.426 (35.4)		
21.03	21.17	0.105	0.517 (22.5)	21.07	50
21.17	22.90	0.017	0.658 (43.5)		
22.90	23.05	0.090	0.374 (24.7)	22.94	60
23.05	23.55	0.085	0.631 (9.9)		
23.55	23.74	0.204	0.328 (24.6)	23.63	120
23.74	24.19	0.066	0.806 (65.6)		
24.19	24.34	0.110	1.100 (9.6)	24.23	80
24.34	25.19	0.070	1.167 (3.5)		
25.19	25.47	0.183	1.032 (5.0)	25.65	350
25.47	25.65				
26.40	26.61	0.197	0.619 (19.9)	26.44	160
27.71	27.89	0.348	0.554 (14.3)	27.81	100
28.23	28.42	0.104	1.250 (10.4)	28.35	110
28.42	29.60	0.024	0.323 (26.9)		
29.60	29.78	0.055	0.943 (70.0)		
30.71	30.88	0.261	0.441 (18.3)	30.91	70
30.88	31.91	0.137	0.518 (7.7)		
31.91	32.13	1.010	0.708 (7.6)	32.10	80
33.31	33.59	0.874	0.401 (9.5)	33.55	70
34.21	34.52	1.260	1.038 (4.5)	34.39	90
34.52	34.83	1.280	0.823 (5.6)		
34.83	35.02	2.060	0.830 (4.9)	34.85	140
35.02	35.32	2.570	1.027 (4.3)	35.21	110
35.32	36.50	0.476	1.326 (3.6)	35.75	490
36.50	36.79	0.191	1.362 (5.4)	36.64	120
36.79	38.20	0.148	1.385 (3.1)		
38.20	38.51	0.342	1.038 (5.4)	38.42	190
39.18	39.54	1.960	1.008 (4.2)	39.44	90
40.48	40.76	0.537	0.675 (8.8)	40.51	150
41.33	41.74	0.828	0.757 (8.3)		
41.74	42.06	0.856	0.693 (6.8)	41.91	80
43.23	43.64	0.500	0.544 (4.0)	43.41	40
43.91	44.16	0.807	0.633 (3.9)	44.04	40
44.51	45.01	1.140	1.060 (4.0)	44.75	150
45.55	46.01	0.342	0.574 (8.1)	45.79	170
46.74	47.29	1.360	0.620 (5.1)	46.93	80
48.00	48.51	1.480	0.769 (4.5)	48.06	120
48.77	49.07	1.090	0.875 (5.0)	48.82	30
49.07	49.44	0.730	0.784 (7.6)		
49.44	49.79	0.677	0.967 (10.9)	49.51	60
50.37	50.79	1.530	0.838 (5.9)		
50.79	51.10	1.760	0.928 (4.7)		
51.10	51.55	3.370	0.760 (4.4)	51.37	130
52.12	52.68	2.380	1.141 (4.0)	52.27	250
53.34	53.73	0.900	1.042 (14.5)	53.56	140
55.41	55.78	2.630	0.816 (5.2)		
56.38	57.07	3.920	0.480 (4.7)	56.58	70
57.07	57.91	1.160	0.572 (6.3)	57.83	100
57.91	58.46	2.200	0.603 (5.4)	58.15	80
58.46	59.13	1.700	1.151 (4.2)	58.68	120
59.13	60.16	0.723	0.723 (5.6)		
60.16	60.96	1.320	0.613 (5.4)	60.25	100
60.96	61.48	1.050	0.649 (8.9)	61.10	120
61.48	63.58	0.502	0.982 (4.0)		
63.58	64.41	0.800	1.145 (4.3)		

TABLE I *(Continued)*

Energy interval E_1 (eV)	E_2 (eV)	Corrected fissions/g (units of 10^9)	Corrected $R_{\mathrm{Cd/Mo}}$ (±% standard deviation)	Major resonance energy[a] (eV)	ν (meV)[b]
64.41	65.17	0.127	1.189 (10.0)		
65.17	66.03	0.131	1.011 (10.0)		
66.03	66.70	0.291	1.085 (16.9)		
66.70	70.34	0.346	1.163 (4.7)		
70.34	71.26	3.810	0.585 (8.9)		
71.26	72.34	1.220	0.713 (10.8)		
72.34	73.11	2.790	0.406 (17.6)		
73.11	74.57	0.585	0.947 (7.4)		
74.57	75.48	2.710	0.607 (11.6)		
75.48	76.14	1.790	0.979 (8.3)		
84.05	85.25	3.150	0.962 (5.7)		
85.25	86.30	1.400	0.868 (11.5)		

[a] See Ref. 4.

[b] Parameters of Ref. 4 were used except for those derived from Reich-Moore-type parameters of Cramer at 36.64 and 46.93 eV as listed in G. de Saussure and R. B. Perez, Oak Ridge National Laboratory Report No. ORNL-TM-2599, 1969 (unpublished).

The autoradiographs of the three 60° metal sectors exposed to neutrons are shown in Fig. 2. Dark bands caused by resonance fission are observed at each of the neutron energies identified in Table I. These bands of exposed metal were cut in narrow radial strips of 0.5–1 g weight, dissolved, and analyzed for ^{99}Mo, ^{111}Ag, and ^{115}Cd. The ratio of 2.3-day ^{115}Cd activity to 2.7-day ^{99}Mo

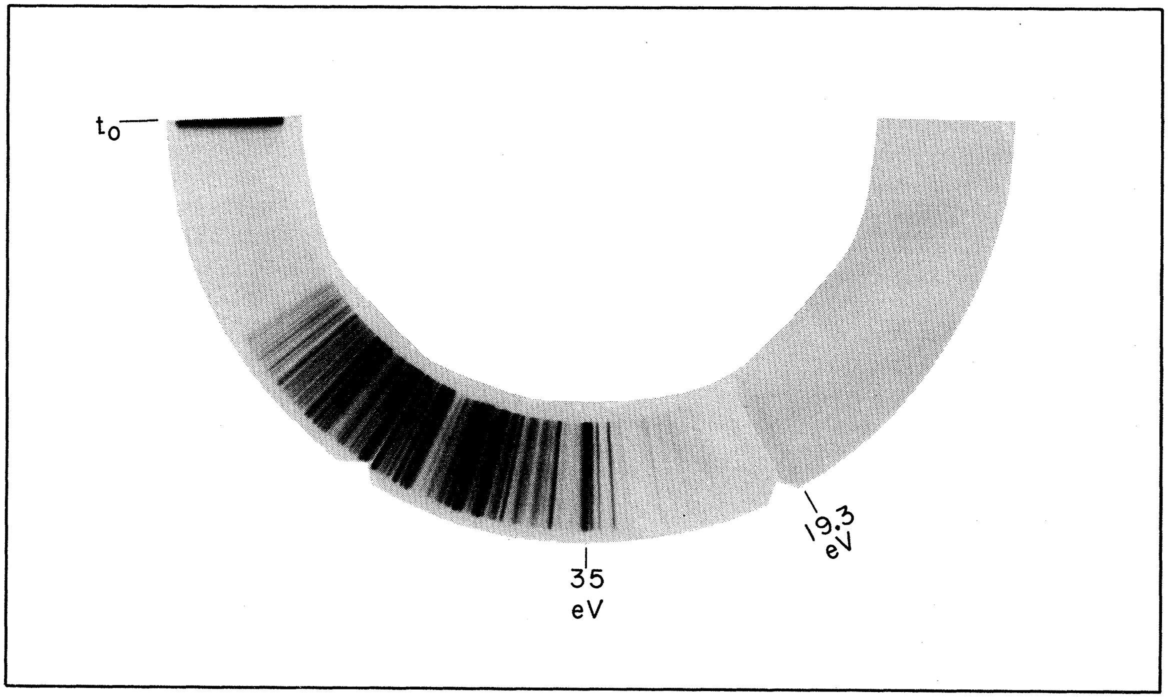

FIG. 2. Autoradiograph of ^{235}U metal produced by contact exposure of x-ray film to fission products in target. An energy scale is provided by identification of t_0 (from γ-ray and fast-neutron-induced fission) and a few prominent resonances.

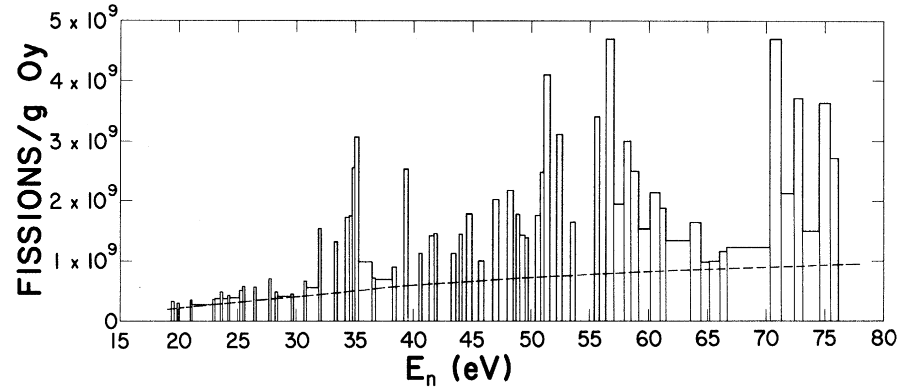

FIG. 3. Experimental results on fission density. Width of bars represents width of metal cuts in eV. The estimated background is shown as a dashed line. "Oy" (oralloy) refers to uranium enriched in ^{235}U.

is normalized to the thermal-neutron value of this ratio and defined as $R_{\mathrm{Cd/Mo}}$. The ^{111}Ag data are converted to corresponding ^{115}Cd equivalents and included in the weighted mean values of $R_{\mathrm{Cd/Mo}}$ which are listed in Table I. Where the values fail to agree within statistics, the result with the smallest statistical error is chosen. We assume that the yield of ^{99}Mo is constant and that changes in the value of $R_{\mathrm{Cd/Mo}}$ are a measure of changes in the yield of ^{115}Cd. The measured fission densities, based on ^{99}Mo analyses, are plotted in Fig. 3. The smoothed estimated background is shown as a dashed line. Part of the background is due to the 0.8–1.0 cm of tapered aluminum wheel immediately behind the target. Another part is due to scattered neutrons in the line of sight. An additional component is due to fission neutrons produced in the target and is enhanced in the neighborhood of large resonances. An additional "background," in a practical sense, is due to the contribution of tails of nearby resonances which may have a different ^{115}Cd yield than the resonance which is being examined. We have not attempted to evaluate this component, partly because a single-level formula is inadequate to make the corrections at these level densities, and the corrections vary widely depending on how a choice is made of resonance parameters from a large number of differing values available from multilevel fits. However, we believe that the smoothed background correction applied to our data is generally accurate to 10–15%. This uncertainty has apparently introduced no significant effect on the qualitative separation of the resonances into characteristic groups.

Values of $R_{\mathrm{Cd/Mo}}$ at resolved resonances are listed in Table II. A few visible resonances are not included in Table II for one or more reasons which are discussed at greater length later. The multilevel fit we have used to correlate with our data is that of Derrien and de Saussure,[4] and their assigned resonance widths ($\nu = \Gamma_{\mathrm{tot}}/2$) are listed in Table I.

The frequency distribution of all corrected values for $R_{\mathrm{Cd/Mo}}$ is plotted in Fig. 4. Two groups are apparent with peak frequencies in the 0.6–0.7 bracket and in the 1.0–1.1 bracket. There are 24 levels in group I, hereinafter referred to as I, and 14 levels in group II, hereinafter referred to as II. The frequency of levels as a function of values of Γ_f (derived from values of ν assigned by Derrien and de Saussure) is also plotted for I and II. It seems evident that these groups have somewhat different distributions in fission width. These distributions resemble those observed for I and II populations identified in the ^{239}Pu measurements of fission symmetry.[1] From Porter-Thomas arguments[5] they indicate that several channels (three or more) are available for fission in both groups.

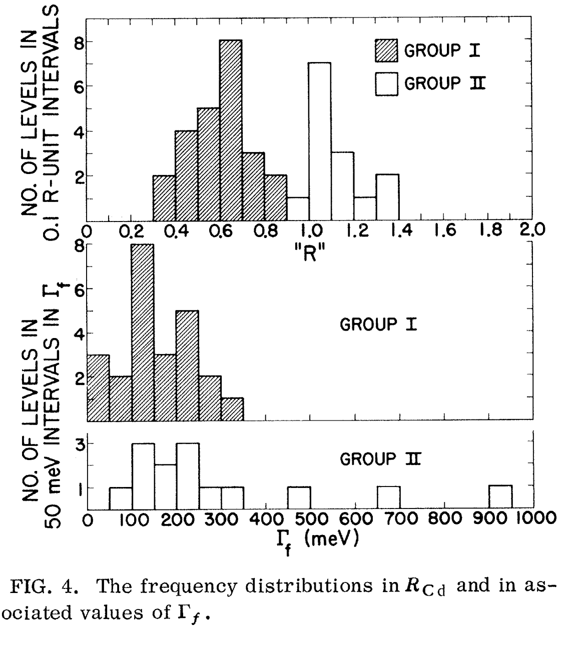

FIG. 4. The frequency distributions in R_{Cd} and in associated values of Γ_f.

The relative populations of levels assigned to I and II are 63 and 37%, respectively. If $J=4$ in I and 3 in II, the prediction for the relative populations is 56.2 and 43.8%, respectively. The average value of Γ_f in I is 146 meV, and in II it is 296 meV. The average value of R in I is 0.593, and in II it is 1.110. These average values can be used to calculate that thermal ^{235}U fission is predominantly of type II (22% of I and 78% of II).

Fission resonances in the 20–60-eV region which are not included in Table II were omitted for one or more of the following reasons:

(1) The resonance is too weak to be seen and was not sampled.

(2) The background correction is 80% or more of the total activity in the analyzed sample.

(3) The resonance is much weaker than a closely adjoining resonance from which it is not resolved.

(4) Two adjoining resonances of comparable size are not resolved from one another and are both excluded.

(5) The sample was not analyzed due to time limitations.

The omitted levels are at the following energies: 20.21, 20.61, 23.43, 27.21, 28.77, 29.68, 30.59, 37.06, 39.92, 41.39, 41.64, 42.16, 42.70, 44.97, 46.98, 48.35, 50.24, 50.45, 51.68, 53.98, 55.16, 55.87, 59.75, and 60.76. The fission widths of the omitted levels average 200 meV compared to an over-all average fission width for the analyzed samples of 201 meV. We conclude that since the

TABLE II. Values of $R_{\mathrm{Cd/Mo}}$ at resolved resonances.

$E_0^{(4)}$ (eV)	$\bar{E}_{\mathrm{sample}}$ (eV)	$R_{\mathrm{Cd/Mo}}$ (±% standard deviation)		Γ_f (meV)[a]
19.30	19.46	0.402	(26.3)	100
21.07	21.10	0.517	(22.5)	60
22.94	22.98	0.374	(24.7)	80
23.63	23.64	0.328	(24.6)	200
24.23	24.26	1.100	(9.6)	120
25.65	25.42	1.032	(5.0)	660
26.44	26.50	0.619	(19.9)	280
27.81	27.80	0.554	(14.3)	160
28.35	28.32	1.250	(10.4)	180
30.91	30.80	0.441	(18.3)	100
32.10	32.01	0.708	(7.6)	120
33.55	33.45	0.401	(9.5)	100
34.39	34.36	1.038	(4.5)	140
34.85	34.92	0.830	(4.9)	240
35.21	35.17	1.027	(4.3)	180
35.75	35.91	1.326	(3.6)	940
36.64	36.54	1.362	(5.4)	200
38.42	38.36	1.038	(5.4)	340
39.44	39.36	1.008	(4.2)	140
40.51	40.62	0.675	(8.8)	260
41.91	41.90	0.693	(6.8)	120
43.41	43.44	0.544	(4.0)	40
44.04	44.04	0.633	(3.9)	40
44.75	44.76	1.060	(4.0)	260
45.79	45.78	0.574	(8.1)	300
46.93	47.02	0.620	(5.1)	120
48.06	48.26	0.769	(4.5)	200
48.82	48.92	0.875	(5.0)	20
49.51	49.62	0.967	(10.9)	80
51.37	51.32	0.760	(4.4)	220
52.27	52.40	1.141	(4.0)	460
53.56	53.54	1.042	(14.5)	240
56.58	56.72	0.480	(4.7)	100
57.83	57.49	0.572	(6.3)	160
58.15	58.18	0.603	(5.4)	120
58.68	58.80	1.151	(4.2)	200
60.25	60.56	0.613	(5.4)	160
61.10	61.22	0.649	(8.9)	200

[a] Γ_f is derived from ν from equation $\Gamma_f = (2\nu) - (40\text{ meV})$.

TABLE III. Summary of level assignments in groups I and II.

Group I E_0 (eV)	Group II E_0 (eV)
19.30	
21.07	
22.94	
23.63	
	24.23
	25.65
26.44	
27.81	
	28.35
30.91	
32.10	
33.55	
	34.39
34.85	
	35.21
	35.75
	36.64
	38.42
	39.44
40.51	
41.91	
43.41	
44.04	
	44.75
45.79	
46.93	
48.06	
48.82	
	49.51
51.37	
	52.27
	53.56
56.58	
57.83	
58.15	
	58.68
60.25	
61.10	

widths of the omitted samples are not noticeably different from those in the population of analyzed samples, the omissions probably do not affect the relative distributions. The level assignments in I and II are summarized in Table III. A tendency for levels in II to cluster is evident.

III. DISCUSSION

The relative populations of levels assigned to I and II are in reasonable agreement with the prediction from $(2J+1)$ statistics if $J=4$ in I and 3 in II. However, we cannot establish a correlation between these assignments and various published sets of ^{235}U spin assignments.[6-8]

On the basis of (1) the fact that in ^{239}Pu the pattern of symmetry and fission width distribution is demonstrated to be related to spin; (2) the measured values of the relative populations of I and II levels in ^{235}U; and (3) the observation that the broader levels occur in II as might be expected if fission in these levels generally occurs through lower-lying channels than in I, we are inclined to favor the hypothesis that $J=4$ in I and 3 in II.

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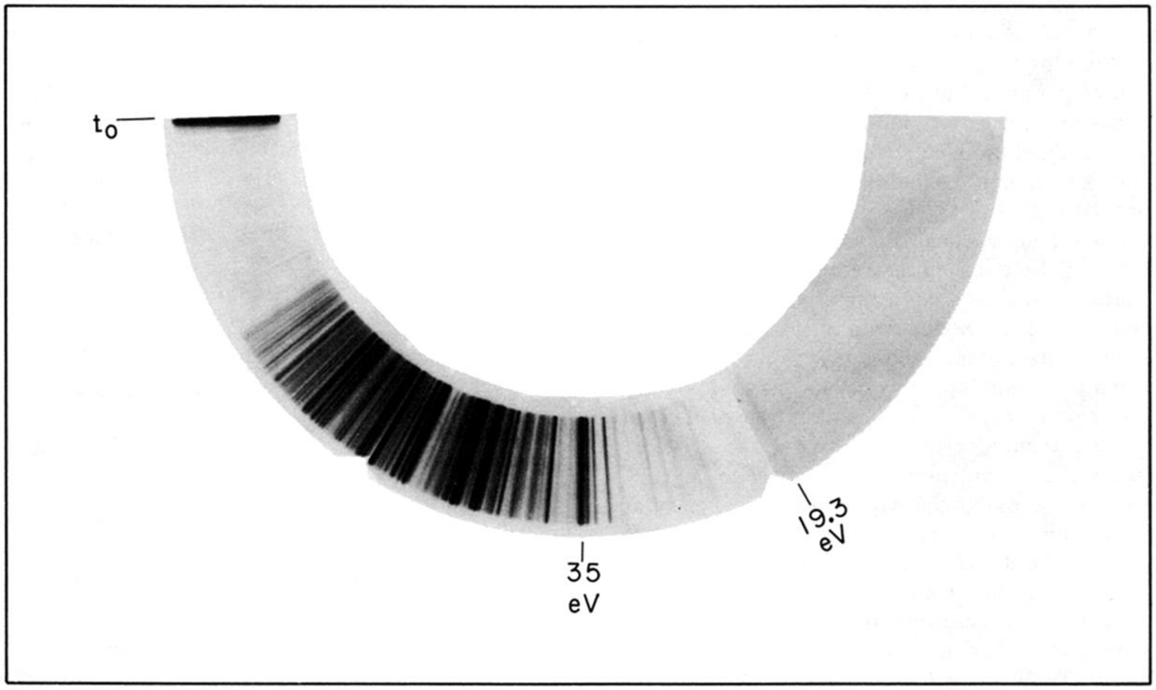

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