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CONVERSION ELECTRON SPECTRA OF Cm^{242} AND Cm^{244}

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ABSTRACT

The conversion electron spectra of Cm^{242} and Cm^{244} have been studied by means of two 180° photographic recording beta-ray spectrographs. Conversion coefficient ratios in the L and M subshells have been measured for several E2 transitions in the decay of these nuclides, and the values so obtained compared with theoretical ratios. Accurate energy determinations of the first three excited states in Pu^{238} are reported, and the validity of the Bohr-Mottelson rotational energy formula, including the vibration-rotation interaction term to describe these states, is discussed. The energy of the first excited state in Pu^{240} is reported. The decay of I^{131} is discussed briefly.

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I. INTRODUCTION

The study of low-energy conversion electron spectra among elements of high atomic number is especially attractive because here the energy separation between atomic orbitals is large and one is able, at moderate resolution, to obtain information regarding internal conversion in the several L, M, and N subshells for a given gamma ray. Such information can be of value not only insofar as the precise measurement of gamma ray energies is always helpful in the solving of complex decay schemes, but also because the determination of the relative probabilities of internal conversion in the various subshells can lead to an elucidation of the multipolarities of the transitions; or, in those cases where theoretical calculations of the conversion coefficient ratios have not yet been made, one can hope that experimental measurements with gamma transitions of known multipolarities will provide some basis for further theoretical work.

The systematic behavior of the excited states of even-even nuclei in the heavy-element region has been a subject of much interest and relevance to current nuclear models; thus it is also desirable to obtain a more detailed knowledge of the energies of these states than is presently enjoyed.

In this paper we report on the following: (1) precision measurements of the energies of the first excited states in Pu^{238} and Pu^{240}, (2) determination of the L, M, and N shell internal conversion ratios for E2 transitions in the above nuclei and comparison with theoretical predictions, and (3) measurement of the energies of the second and third excited states in Pu^{238} and a comparison with the energies expected for these states from the Bohr-Mottelson rotational model.

II. INSTRUMENT DESCRIPTION

Two flat 180° spectrographs of the permanent magnet type have been constructed, which employ photographic recording of the conversion lines. Mihelich[1] and Slätis[2] have pointed out the advantages of this type of instrument for conversion electron studies: (a) moderately high resolution is readily available, (b) very good stability of the field is obtained, provided that the magnet temperature is held constant, and (c) the photographic film records line positions accurately and integrates the entire spectrum simultaneously, providing a permanent

record of primary data. Disadvantages of this method are principally that: (a) photographic film is an extremely nonlinear exposure recording device and can be used for obtaining relative intensities of lines only after extensive calibrations and (b) to approach optimum resolution with these instruments one needs thin sources deposited on fine wires, a condition which is often difficult to meet in practice.

The two spectrographs used in this work have field strengths of ~53 gauss and ~99 gauss, respectively. Their vacuum chambers and cameras are a modification of the Brookhaven design[3] and are shown in Fig. 1. The maximum radius of curvature is about 20 cm, which corresponds to maximum electron energies in the two instruments of ~80 and ~275 kev, respectively. No provision is made in the cameras for rapid source placement or film removal because only relatively long-lived isotopes are being studied at the present time.

Eastman No-Screen X-ray film on glass backing 3/4 x 15 x 0.04 inch has been used to record the lines. The calibration of this film is described below.

III. SOURCE PREPARATION

The spectrograph sources were prepared by electrodeposition of the active materials upon 10-mil or 14-mil platinum wires using a

procedure suggested by B. G. Harvey, in which $M(OH)_3$ (where M is the actinide cation) is deposited on the wire cathode of an electrolysis cell using NH_4HSO_4 at a pH of 3.6 as the electrolyte. A drawing of the electrolysis cell is shown in Fig. 2. The active wires are mounted in a special source holder, shown in Fig. 3, whose position in the camera could be adjusted and reproduced within several mils.

IV. INSTRUMENT CALIBRATION

Calibration of the spectrographs consisted in the determination of the effective magnetic field at various ρ values, using accurately known gamma ray energies of I^{131} and Am^{241} and conversion line energies of ThB. The energies used for calibration are given in Table I.

TABLE I

Calibration Standards[4]

Standard	Energy or $H\rho$	Reference
ThB	A line, $H\rho$ = 534.11	5
	F line, $H\rho$ = 1388.5	5, 6, 7
	I line, $H\rho$ = 1754.0	5, 6, 7
I^{131}	80.164 kev	8
	284.307 kev	8
Am^{241}	33.20 kev	9
	43.46 kev	9
	59.57 kev	9

Figure 4 shows the calibration curve which has been obtained for the 53-gauss spectrograph. The effective field in this instrument is known to about 0.1 percent from $\rho = 9$ cm to $\rho = 19$ cm; the field in the 99-gauss spectrograph is known to a similar accuracy.

As part of the calibration procedure, a series of accurately machined notches was cut into the film holder so as to render a permanent set of light and dark reference edges exposed into each photographic plate. With this device, similar to that described by Slätis,[2] the notch distances and other dimensions pertinent to a determination of ρ are all constants of the spectrograph camera and need be measured only once. In subsequent experiments only the distance of each electron line to the nearest notch shadow need be measured; in no case will this distance be greater than 2 cm, the distance between notch edges. For the energy determinations we have used an ordinary x-ray diffraction film comparator, with which the distances could be measured reproducibly to ±0.01 cm.

The resolution $\Delta\rho/\rho$ obtained in the 53-gauss spectrograph for the I^{131} (80 kev) K line is 0.17 percent and for the L_I line 0.13 percent; in the 99-gauss spectrograph the best resolution has been 0.17 percent. The instruments are located in a room whose temperature variation is only a few degrees Fahrenheit; better thermostating is being planned, so that loss in resolution due to temperature variation will be minimized.[2]

V. FILM CALIBRATION AND RELATIVE INTENSITY DETERMINATIONS

In order to determine relative intensities of lines one must study several features of the photographic action of electrons.

The photographic density as a function of exposure (number of electrons striking the film) must be determined experimentally for the particular film being used. Since the shape of the D vs E curve has been shown to be independent of electron energy,[10,12] we have used the beta spectrum of Sr^{90} - Y^{90} for this purpose. Our experimental D vs E curve is shown in Fig. 5.

The dependence of line intensity on the radius of curvature of the electron path must be determined; Slätis[2] has shown that the intensity is inversely proportional to ρ providing the collimating slit is in the plane of the film. Our spectrographs have been designed to fulfill this condition.

The photographic blackening efficiency as a function of electron energy must be ascertained. Cranberg and Halpern[11] measured this efficiency for No-Screen film in the range of 9 to 40 kev with the use of an electron gun, and Dudley[12] covered the range 35 to 1800 kev using radioactive sources. The slopes of the efficiency curves given by these authors were approximately the same in the region of overlap; therefore, the curves were normalized and the resultant curve was used for our intensity measurements. This curve is shown in Fig. 6.

The relative intensity of an electron line is given by

$$I = A\rho/\eta$$

where A = area under line, ρ = radius of curvature, and η = efficiency of film. In our experiments, the area A (as obtained from a densitometer plot of the blackening) was determined by the method of Mladjenovic and Slätis,[13] which gives a good approximation to the true area of the line with a great saving of labor. In essence, the following is done: The D vs E calibration curve is used in conjunction with the densitometer plot of the experimental film to establish the maximum height of each line in arbitrary units of electron density and also to determine the point of half-maximum density in the same units. At this half-way point the line width is measured, and the area of the rectangle determined by the line width and height is taken as an approximation to the true area of the electron line.

All photometer measurements were made with an Applied Research Laboratories densitometer connected to a L and N Speedomax Recorder.

VI. EXPERIMENTAL RESULTS

A. Iodine-131

I^{131} was used primarily as a calibration standard, but some observations made with very active sources of this isotope are perhaps worth reporting.

In addition to the K, L_I, M_I and N lines of the well-known 80.16-kev transition, we have observed in low intensity the L_{II} line of this transition, previously unreported. Its intensity,

relative to that of the L_I line, appears from the densitometer tracing to be roughly 0.10. This figure had previously been given as an upper limit by Mihelich. This transition is known to be a magnetic dipole, and the theoretical calculations of Rose et al.[14] predict an L_{II}/L_I ratio (for Z = 55, E = 75 kev) of 0.07.

Three other novel lines were also seen. These were the K and L_I lines of a 163.8 ± 0.1-kev transition, and the K line of a 177.1 ± 0.1-kev transition. The 164-kev level has been assigned to the 12-day Xe^{131m_2} isomeric level, and its energy was previously determined by Bergström[15] to be 163.9 kev. The 177-kev K line has been sporadically reported[16,17] but it does not fit into the presently accepted[18] decay scheme of I^{131}.

B. Curium-242

The energy of the first excited state of Pu^{238}, populated by the alpha decay of Cm^{242}, has been measured in several ways. O'Kelley[19] and Passell[20] using a double-focusing beta spectrometer observed conversion electrons corresponding to a gamma ray of 44.9 kev. Asaro et al.[21] using a magnetic alpha spectrograph have studied the complex alpha spectrum of Cm^{242} and have found an energy difference between the most prominent alpha groups which corresponds to an energy of 44.3 kev for the first excited state of Pu^{238}. More recently, Slätis, Rasmussen, and Atterling[22] have studied the beta decay of Np^{238} using a double-focusing spectrometer at 0.5 percent resolution and have found the energy of the first excited state of Pu^{238} to be 44.1 ± 0.1 kev.

Our present data, from four separate one-day exposures in the 53-gauss spectrograph, are given in Table II. From these experiments a best value for the energy of the first excited state of Pu^{238} is given as 44.11 ± 0.05 kev. This transition, like all transitions from the first excited state to ground state in even-even nuclei, is of multipole order E2. It has been so characterized by measurements of the total conversion coefficient[21] and from the L subshell ratios. Previously reported values for the L_{II}/L_{III} intensity ratio as studied from the beta decay of Np^{238} are Freedman et al.[23] 1.91, Passell[20] 1.44, and Slätis et al.[22] 1.37. From the decay of Cm^{242}, Passell[20] obtained a value of 1.26.

We have measured the L and M subshell ratios for this transition and have estimated the N ratios. A densitometer plot of one of the Cm^{242} films is shown in Fig. 7 together with a linear retracing made with the aid of the empirical D-E curve. The relative intensity determinations, which are summarized in Table III, indicate that the L_{II}/L_{III} and M_{II}/M_{III} ratios for this transition are identical, to within about 10 percent, and have the value 1.2 ± 0.1. M_{IV} and M_V conversion is not observed, and M_I conversion is extremely weak.

TABLE II

Energies of Cm^{242} Conversion Electrons

Exp. No.	Standard on same wire	Conversion Subshell and Electron Energies							
		L_{II}	L_{III}	M_{II}	M_{III}	N_{II}	N_{III}	O_{III}	M_{I} (?)
a	I^{131}	21.85	26.05	38.55	39.55	42.73	43.00	--	--
b	I^{131}	21.87	26.06	38.60	39.62	42.75	43.01	43.90	38.23
c	ThB	21.86	26.05	38.56	39.55	42.70	42.95	43.85	--
d	--	21.87	26.04	38.53	39.56	42.70	42.96	43.88	--
Average Electron Energy		21.86	26.05	38.56	39.57	42.72	42.98	43.88	--
Binding Energy		22.25	18.06	5.56	4.56	1.38	1.13	0.23	5.93
Transition Energy		44.11	44.11	44.12	44.13	44.10	44.11	44.11	44.16

TABLE III

Relative Intensities of Conversion Lines from 44.1-Kev Transition of Cm^{242}
Theoretical Value: $L_{II}/L_{III} = 1.2$

Exp.	L_{II}/L_{III}	M_{II}/M_{III}	N_{II}/N_{III}	$L_{\Sigma}/M_{\Sigma}/N_{\Sigma}$
A	1.23	1.25	~1	2.5/1.0/0.3
B	1.27	1.22	~1	2.9/1.0/0.3
C	1.16	1.20	~1	2.4/1.0/0.3

Our M ratios should be somewhat more reliable than our L ratios for this transition because the parameters ρ and η remain practically constant over the small energy range of the M lines and hence any uncertainty in their absolute values will cancel out in the ratio; however, in the region of the L electron energies the slope of the efficiency-energy curve is steep and therefore the accuracy of the L_{II}/L_{III} ratio depends strongly on the accuracy of this calibration. The N_{II} and N_{III} lines were not completely resolved, but their intensities were estimated visually to be about equal.

The theoretical L_{II}/L_{III} ratio given by Gellman et al.[24] for an E2 transition of 44 kev in Z = 92 is 1.2. The L_{II}/L_{III}

ratio obtained in these experiments is in good agreement with their theoretical value. There are at present no theoretical calculations of M subshell conversion coefficients with which to compare the experimental results other than the "threshold" calculations of Church and Monahan[25] which state that E2 radiation converts primarily in the M_{II} and M_{III} shells. It appears that such calculations would be of value especially in the study of higher multipoles where M_{IV} and M_V conversion becomes important.

It was desirable to study also the conversion lines from the higher excited states of Pu^{238}. Since the population of the second and third excited states from the alpha decay of Cm^{242} is only 0.035 percent and 0.009 percent, respectively,[21] an exposure of one month's duration was taken in the 99-gauss spectrograph in an attempt to record these weak lines. Although many "extra" lines appeared on this plate due to very small amounts of Cm^{243} and Am^{241} in the sample, a series of lines could be assigned to a 101.9 ± 0.3-kev transition and a 157.7 ± 0.5-kev transition in Pu^{238}. The electron energies and their assignments are given in Table IV, and the level scheme is shown in Fig. 8.

We have measured the relative intensities of the subshell electron groups from the 101.9-kev transition, with the result $L_I/L_{II}/L_{III}$ = ~0.2/1.4/1.0. These ratios confirm the predominantly E2 nature of this transition and check with the theoretical values of Gellman et al.[24] for E = 100 kev and Z = 92, which are $L_I/L_{II}/L_{III}$ = 0.14/1.8/1.0. The subshell ratios indicate an

upper limit for possible MI mixing of about 15 per cent. As was the case with the 44-kev transition, the M subshell ratios are here similar to the L ratios; the M lines were weak, but a rough value is $M_{II}/M_{III} = 1.5/1.0$. Also $L_{\Sigma}/M_{\Sigma} \approx 3$.

The excited states of Pu^{238} form a band which is characteristic of the heavy even-even nuclides.[26] These levels are described by the unified nuclear model of Bohr and Mottelson[27] as rotational states arising from surface excitations of a deformed nucleus. The configurations expected are I = (0+), (2+), (4+), etc., and the energies are approximately equal to $(\hbar^2/2\mathfrak{J})I(I + 1)$ where $\mathfrak{J}$ is the effective moment of inertia of the deformed nucleus. Deviations from this strict $I(I + 1)$ dependence are predicted[27] for the higher excited states where centrifugal distortions slightly increase the equilibrium nuclear deformation; the correction term is of the form $\Delta E = -2(1/\hbar\omega)^2 (\hbar^2/\mathfrak{J})^3 I^2 (I + 1)^2$ where $\hbar\omega$ is the vibrational quantum.

The measurements reported here for the energies of the (2+), (4+), and (6+) states in Pu^{238} allow two independent calculations to be made of the constants A and B in the Bohr-Mottelson equation $E = AI(I + 1) - BI^2(I + 1)^2$. From the energies of the (+2) and (+4) states one has A = 7.37 kev and B = 0.0035 kev; from the (2+) and (6+) states one finds A = 7.37 kev and B = 0.0033 kev. This close agreement, unless fortuitous, indicates that the Bohr-Mottelson formulation accurately describes the energies of the first four rotational states in Pu^{238}. With the above constants, the equation may be used to predict the energy of the fifth rotational

state, the (8+) state, to be at 513.1 kev. It is interesting that F. Asaro and I. Perlman[28] have recently found weak gamma radiation in Cm^{242} at 210 ± 10 kev which is in good agreement with the expected (8+) → (6+) transition energy, 513.1 - 303.7 = 209.4 kev.

C. Curium-244

A sample of Cm^{244} containing a few percent Cm^{242} was plated onto a 10-mil platinum wire, and several exposures were made in the 53-gauss spectrograph. The energies of the electron lines so obtained are summarized in Table V and indicate that the first excited state of Pu^{240} has an energy of 42.88 ± 0.05 kev. The Cm^{242} in this sample served as an internal standard.

Relative intensities for L, M, and N subshell conversion were measured, with the results given in Table VI. These results are essentially the same as obtained with Cm^{242} and again support the theoretical L_{II}/L_{III} ratios of Gellman *et al.*[24] for an E2 transition. L_I and M_I conversion lines were observed but were so weak that their relative intensities could not be measured quantitatively.

TABLE IV

Conversion Electrons from Decay of Second- and Third-Excited States of Pu^{238}

Electron energy	78.8	79.6	83.8	96.3	97.3	100.7	135.5	139.6
Relative intensity	~0.2	1.4	1.0	--	--	--	--	--
Subshell	L_I	L_{II}	L_{III}	M_{II}	M_{III}	$N_{II} + N_{III}$	L_{II}	L_{III}
Binding-energy energy	23.1	22.3	18.1	5.6	4.6	1.2	22.3	18.1
Transition energy	101.9	101.9	101.9	101.9	101.9	101.9	157.8	157.7

TABLE V

Energies of Cm^{244} Conversion Electrons

Exp. No.	Electron Energies								
	L_I	L_{II}	L_{III}	M_I	M_{II}	M_{III}	N_{II}	N_{III}	O
a	19.76	20.60	24.80	36.93	37.31	38.28	41.49	41.75	42.63
b	--	20.64	24.84	--	37.36	38.35	41.57	41.79	--
c	19.77	20.60	24.80	--	37.32	38.29	41.49	41.74	42.65
Average Electron Energy	19.77	20.61	24.81	36.93	37.33	38.31	41.52	41.76	42.64
Subshell Binding Energy	23.10	22.25	18.06	5.93	5.56	4.56	1.38	1.13	~0.23
Transition Energy	42.87	42.86	42.87	42.86	42.89	42.87	42.90	42.89	42.87

TABLE VI

Relative Intensities of Conversion Lines from 42.9-Kev Transition of Cm^{244}

Theoretical Value: $L_{II}/L_{III} = 1.2$

Exp.	L_{II}/L_{III}	M_{II}/M_{III}	N_{II}/N_{IV}	$L_{\Sigma}/M_{\Sigma}/N_{\Sigma}$
A	1.08	1.15	~1	2.5/1.0/0.3
B	1.13	1.06	~1	2.4/1.0/0.3

VII. ACKNOWLEDGMENTS

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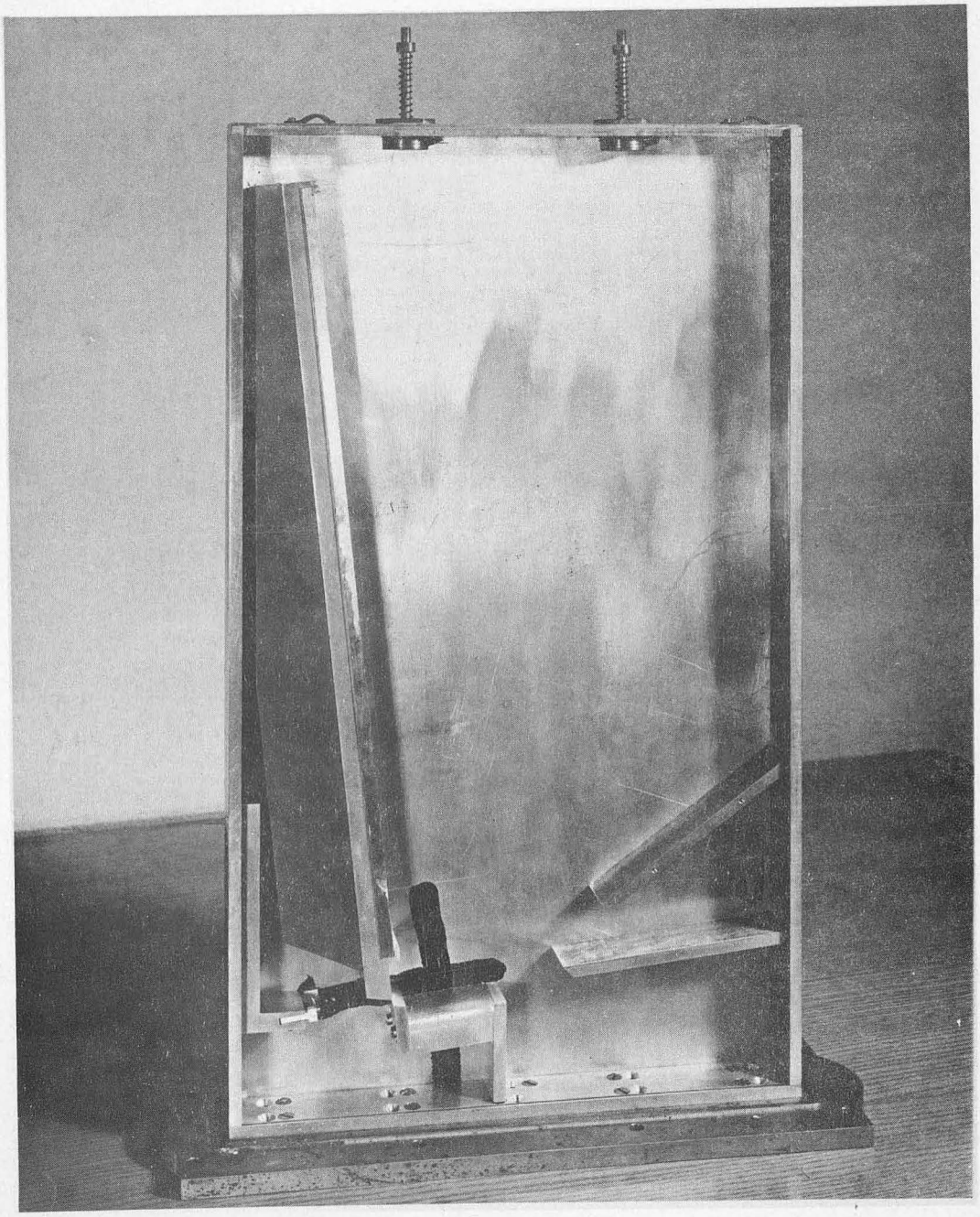

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Fig. 1. Photograph of top view of camera, cover plate removed.

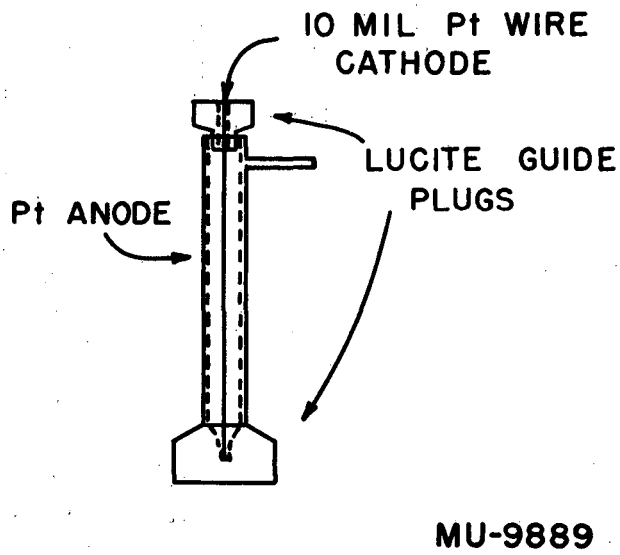

Fig. 2. Drawing of electrolysis cell used for preparing wire sources.

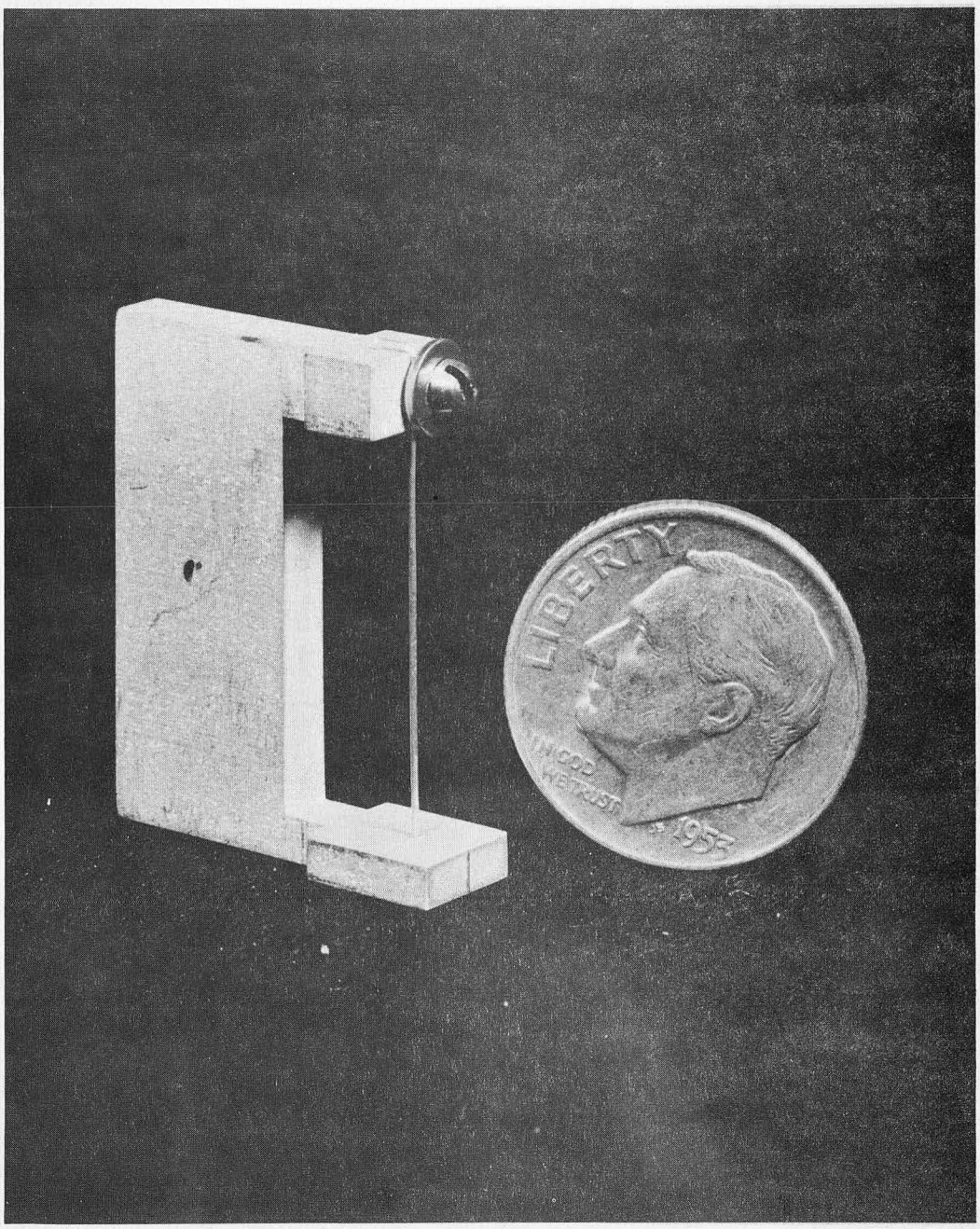

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Fig. 3. Photograph of wire source holder.

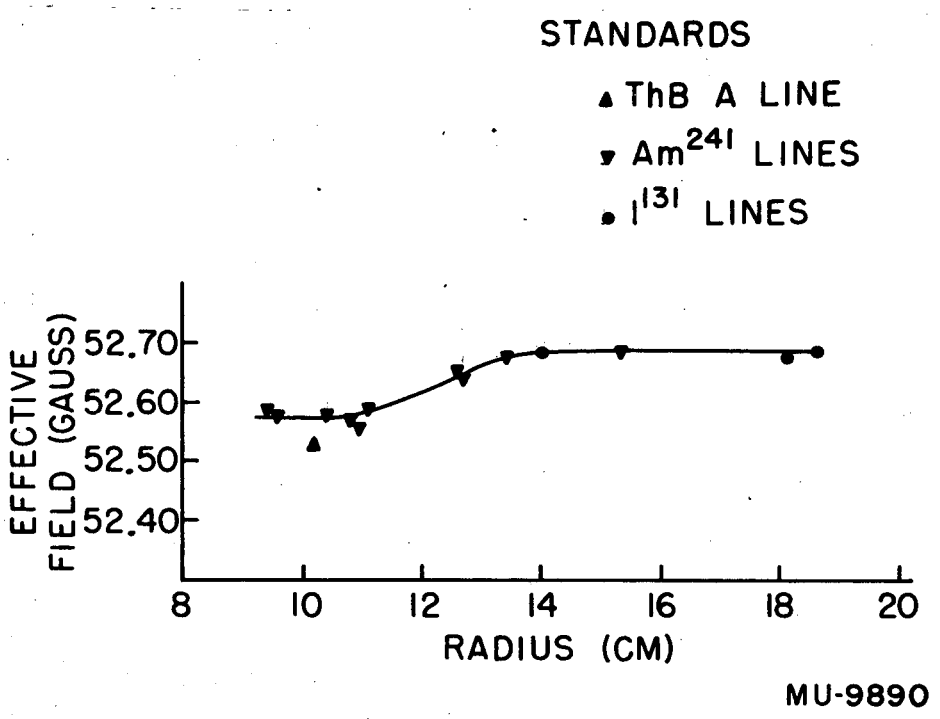

Fig. 4. Calibration curve for 53-gauss spectrograph.

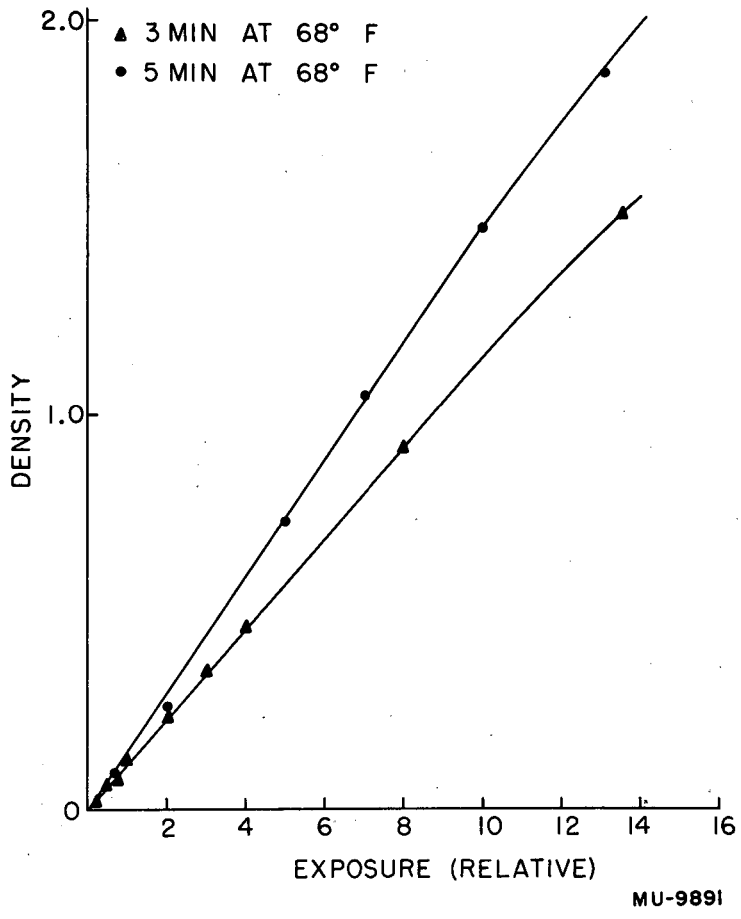

Fig. 5. Photographic density vs exposure time. Eastman No-Screen Film, Sr^{90} - Y^{90} source, D-19 full strength.

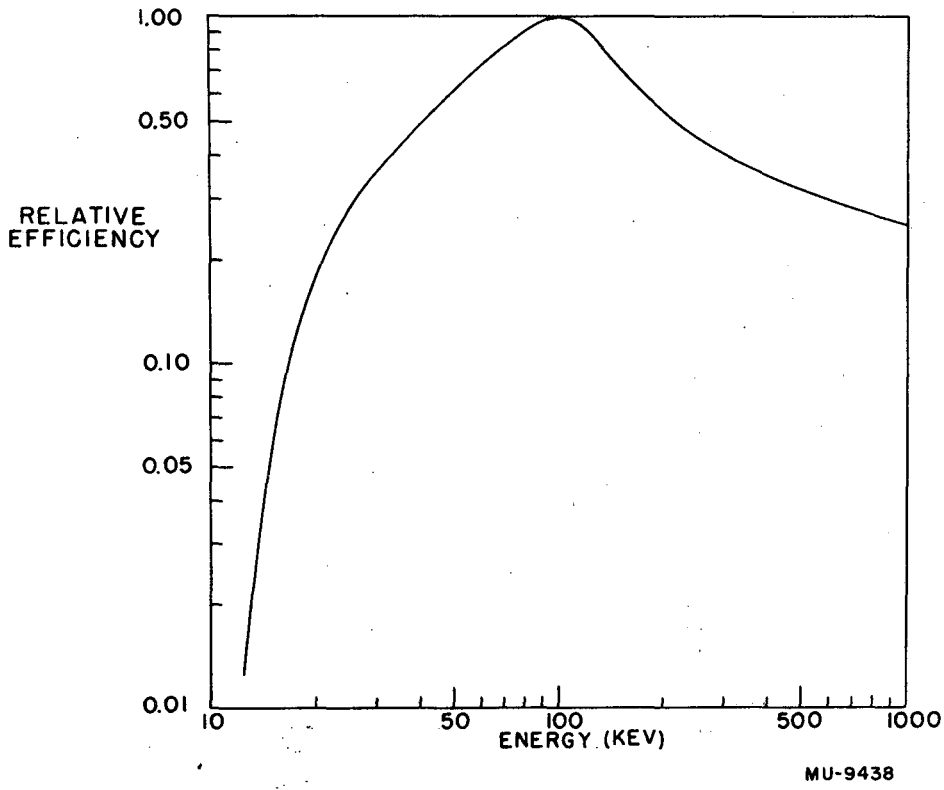

Fig. 6. Photographic efficiency vs electron energy (kev).

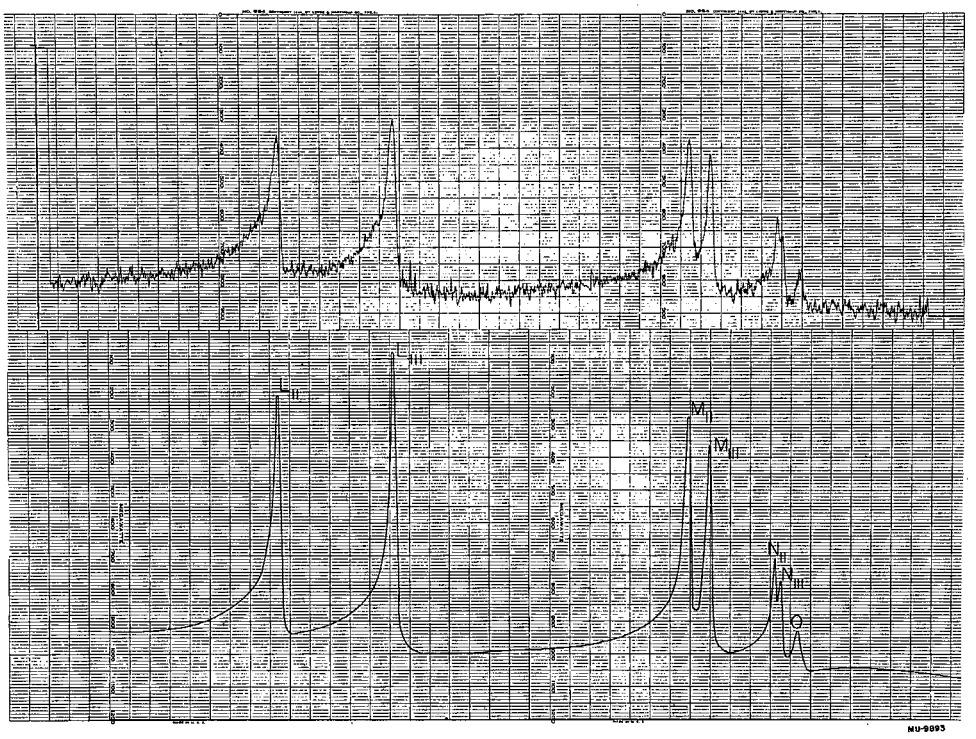

Fig. 7. Densitometer plot of Cm^{242} lines.

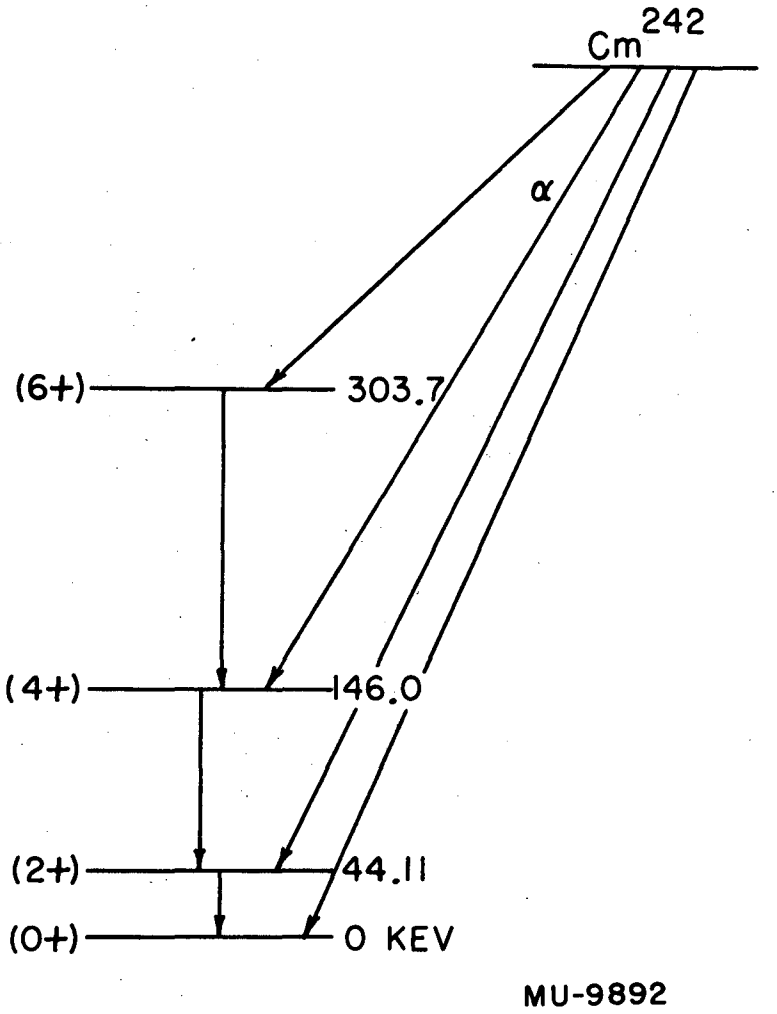

Fig. 8. Cm^{242} decay scheme.