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ABSTRACT

A new gadolinium isotope decaying by electron capture with a 29-hour half-life was found. Its mass number was determined to be 147 by examination of its excitation function for production by alpha particle bombardment of $Sm_2^{147}O_3$. The electron-capture isotope Gd^{149} was also studied, and its half-life was redetermined as 9.3 ± .3 days. The conversion-electron and photon spectra of Gd^{147} and Gd^{149} were studied. Relative photon and electron intensities were determined for some of the transitions, and coincidence measurements were made. From the data a few multipolarities are assigned and partial decay schemes are suggested.

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INTRODUCTION

No previous electron or gamma spectroscopic work on any gadolinium isotopes lighter than Gd^{151} has been reported up to this time. The region around the light gadolinium isotopes is of special interest because of the many sudden changes in nuclear properties seen in passing between 88 and 90 neutrons. From optical spectroscopic results, Brix and Kopfermann[1] have called attention to sharp breaks in isotope shifts occurring between isotopes having 88 and 90 neutrons. They have also noted[2] the large difference in the quadrupole moments of the stable europium isotopes, which have 88 and 90 neutrons. The first excited states of even-even nuclei drop sharply at 90 neutrons, and well-developed nuclear rotational bands appear. The large deformation approximation of the Bohr-Mottelson model[3] has been successful in describing nuclei in this region of $N \geq 90$ but not in the region of $N \leq 88$.

In this work information on the excited levels in Eu^{147} and Eu^{149} was obtained by studying the conversion electron and photon spectra of gadolinium activities produced by alpha particle bombardments on samarium oxide and deuteron bombardments on europium oxide.

EXPERIMENTAL PROCEDURES

Bombardments:

Activities were produced in the Berkeley 60-inch cyclotron by alpha particles accelerated to 48 Mev and deuterons to 24 Mev. The samarium and europium target materials were bombarded in the oxide form in "boats" stamped in 10-mil platinum foil with 1/4-mil platinum foil covers. These "boats" fit into standard target assemblies, and were cooled by water directed at the back of the targets.

Chemistry:

The platinum boats containing the irradiated rare earth oxides were placed in hot 8 M hydrochloric acid until the oxides dissolved. The platinum was then removed and ammonia was passed into the acid solution until the rare earths precipitated as hydroxides. The precipitate was washed and dissolved in a minimum amount of 8 M hydrochloric acid. The gadolinium produced was separated from the remaining rare earths by an ion-exchange method identical to that described by Thompson, Harvey, Choppin, and Seaborg.[4] The column (3 mm in diameter, 5 cm in length) was heated to 87°C by a trichloroethylene vapor jacket. The eluting agent was 0.4 M alpha-hydroxyisobutyric acid buffered to a pH of 3.93 with ammonium hydroxide, and the resin was Dowex-50 spherical resin, 200-400 mesh size, 12% cross-linked. Since the maximum capacity of the column was ~10 mg of rare earths, it was necessary to remove the bulk of the target material by another method. A sodium amalgam reduction method was used for this purpose. Sodium amalgam is capable of reducing tripositive rare earth ions to the metal and amalgamating them; and since both samarium and europium have dipositive states to act as intermediate states, the reduction of Sm^{+3} and Eu^{+3} is much faster than that of Gd^{+3}. The rare earths, dissolved in a minimum amount of 8 M hydrochloric acid, were transferred to a separatory funnel, and 10 ml of water and 8 drops of glacial acetic acid (to maintain the hydrogen ion concentration) were added. 0.3% sodium amalgam (15 ml for 150 mg rare earths) was added and the mixture shaken for 10 seconds. After the amalgam layer was removed, the remaining aqueous layer seemed to retain about 10% of the original target material. The procedure was repeated three times in order to be sure that the total rare earth mass retained was under 10 mg. Ammonia was passed into the final aqueous layer until the rare earths precipitated as hydroxides. The precipitate was washed, dissolved in a minimum amount of 1 M hydrochloric acid, and finally concentrated into 2 drops of 0.05 M hydrochloric acid for adsorption on the column. After completing the ion-exchange separation of the remaining rare earth mass, the drops of alpha-hydroxyisobutyric acid solution containing the gadolinium activity were combined and made 0.5 - 1 M in hydrochloric acid. This solution was placed on a Dowex-50, 4% cross-linked,

3 mm x 5 cm column at room temperature. 0.5 M hydrochloric acid was passed through until all of the alpha-hydroxyisobutyric acid was removed, and the activity was then eluted with 8 M hydrochloric acid.

Instruments:

An argon-flow-type ionization chamber was used for alpha counting. The instruments used for measuring the conversion-electron spectra of the gadolinium activities were four permanent-magnet 180° spectrographs described by Smith and Hollander.[5] They have fields of approximately 50, 100, 200, and 350 gauss and operate at resolutions of about 0.15%. Gadolinium sources for them were plated onto 10-mil platinum wires from a solution of 0.1 M ammonium bisulfate at a pH of 3.6. The procedure used was that suggested by Harvey et al.[6] in which the wire is made the cathode of an electrolysis cell. Since the hydroxide ion concentration around the cathode is high, the rare earths are deposited onto the wire as hydroxides. The wire is held in a bunsen burner flame for a few seconds before placing it in the spectrograph. Electrons from the source impinge on a photographic plate. By previous calibration of the instruments with electrons of known energies the energies of conversion electrons may be determined accurately. To measure the relative intensities of the electrons, densitometer traces of the spectrograph plates were made. Relative intensities of conversion electrons were also measured with more accuracy with a double-focussing spectrometer.[7] This is a shaped-field, 256°, prismatic spectrometer operating at about 0.3% resolution. A 100-channel gamma analyzer[8] with about 8.5% resolution was used to measure gamma-ray energies and intensities. This instrument is a gamma pulse-height analyzer used with a scintillation counter with a 1" x 1-1/2" sodium iodide crystal and photomultiplier tube. It records data by means of a magnetic-core matrix memory, and is able to record at relatively high counting rates.

CONVERSION ELECTRON SPECTRA OF Gd^{147} AND Gd^{149}

From bombardments of natural samarium (3.16% Sm^{144}, 15.07% Sm^{147}, 11.27% Sm^{148}, 13.84% Sm^{149}, 7.47% Sm^{150}, 26.63% Sm^{152}, and 22.53% Sm^{154})[9] with alpha particles, one would expect to make a variety of gadolinium isotopes. Among the unstable of these are Gd^{148} (140-year alpha),[10]

Gd^{149} (9-day electron capture alpha),[9] Gd^{150} (long-lived alpha),[9] Gd^{151} (150-day electron capture),[9] and Gd^{153} (236-day electron capture).[9] In addition, one might expect to produce the unreported isotope, Gd^{147}, by the reactions Sm^{144} (α,n) Gd^{147} and Sm^{147} $(\alpha,4n)$ Gd^{147}.

Using the permanent-magnet spectrographs, electron lines were studied relative to the lines for the 150-kev transition, which was known to decay with a 9-day half-life from gamma analyzer work. Hoff, Rasmussen, and Thompson[11] reported a half-life of 9 ± 1 day for Gd^{149}. We have remeasured it more accurately as 9.3 ± .3 days by following the decay of the conversion electrons of the 150-kev transition. All lines where the energy differences between the conversion-electron energies corresponded to europium binding-energy differences were assigned to gadolinium activities. Of these, all lines the intensities of which stayed the same relative to the intensities of the lines of the 150-kev transition were assigned to Gd^{149}. A second set of gadolinium lines with a half-life of approximately 1-1/2 days (later redetermined as 29 hours) was tentatively assigned to Gd^{147}. The rest of the gadolinium lines were longer-lived. Those resulting from the decay of Gd^{151} or Gd^{153} could be positively identified by seeing if they showed up in a deuteron bombardment designed to produce only Gd^{151} and heavier gadolinium isotopes. In this experiment natural europium, which is composed entirely of Eu^{151} and Eu^{153}, was bombarded with 24-Mev deuterons. A few remaining long-lived gadolinium lines, which were seen in the alpha-particle bombardments of Sm_2O_3 but not in the deuteron bombardments of Eu_2O_3, could not be assigned to the decay of any isotope. These lines may belong to an isomer of one of the gadolinium isotopes, but further study is needed. Lines appearing in later plates which did not appear in earlier ones were assigned to $Eu^{147\text{-}149}$ if, in addition, the energy differences between the conversion electron energies corresponded to samarium binding-energy differences. Eu^{147} has a half-life of 24 days,[9] Eu^{149} one of about 120 days,[9] and Eu^{151} and Eu^{153} are stable.

Table I lists the transitions assigned to the various isotopes with the lines seen for each. The intensities given are visual ones read from the permanent-magnet spectrograph plates. Energies are given to the nearest 0.1 kev if five or more different energy determinations were made and to the nearest 1 kev if less than five were made.

Table I

	Transition Energy (kev)	Lines seen	Visual Relative Intensities* K	L_I	L_{II}	L_{III}	M_1	N_1
Gd^{147}	136	K	VVW					
	142	K	W-M					
	147	K	W					
	217	K	VVW					
	226	K	VVW					
	229.5	K L_I	VVS	W				
		M_1 N_1					VW	VVW
	241	K	VW					
	261.0	K L_1	VW	VVW				
	310	K	VW					
	348	K	VVW					
	370	K L_1	W	VVVW				
	374	K	VVW					
	391	K	VVW					
	396.2	K L_1 M_1	M-S	W			VVW	
	485	K	VVW					
	502	K	VVW					
Gd^{149}	107	K	VVW					
	120	K	VW					
	126.1	K L_I	W-M	W				
	132	K	W-M					
	149.9	K L_1 L_{II}	VVS	VVS	VW			
		L_{III} M_I N_I				VVW	W-M	W
	244	K L_1	VVVW	VVVW				
	253	K	VVVW					
	272.7	K L_1-II	W		VVW			
		M_1					VVW	
	298.8	K L_I	W-M	W				
	346.7	K L_I	VS	W				
		M_I N_I					VW	VVVW
	461	K	VVVW					
	496	K L_I-II	VVW		VVVW			
	517	K	VVW					
	534	K	VVW					
	750	K L_1	VVVW	VVVW				
	790	K	VVVW					
$Gd^{?}$	22.9	L_I L_{II} L_{III}		M	W	W		
		M_I N_I					M	VVW
	77.7	K	VVW					
	114.7	K L_I L_{II}	VS	M	VVVW			
		L_{III} M_I N_I				masked	VVW	VVW
	115.5	K L_I L_{II}	VS	M	VVW			
		M_I N_I					VVW	VVW
	154.8	K L_I	M-S	W				
		M_I N_I					VW	VVVW
	198.8	K L_I	W	VVW				
	329	K	VVW					

(continued)

Table I - continued

Transition Energy (kev)	Lines seen	Visual Relative Intensities* K	L_I	L_{II}	L_{III}	M_I	N_I
$Eu^{147-149}$							
38	L_I L_{III} N_I		VVW		VVW		VVVW
53	L_I L_{III}		VVW		VVW		
66	K	VW					
68	K	M					
69	K	VW					
121.3	K L_I	M-S	VW				
	M_I N_I					VVW	VVVW
219.8	K L_I	VVW	VVVW				
277	K L_I	W	VVVW				

*S = strong, M = moderate, W = weak, V = very.

EXCITATION FUNCTIONS FOR ALPHA-INDUCED REACTIONS ON Sm^{147}

Samarium oxide enriched to 85 atomic percent in Sm^{147}, [12] with the other 15 percent chiefly Sm^{146} and Sm^{149}, was bombarded with alpha particles in a Faraday cup assembly. A stacked-foil technique was used so that excitation functions could be determined. The function for the Sm^{147} $(\alpha,3n)$ Gd^{148} reaction was obtained by alpha-counting the eight target samples, and the function for the Sm^{147} $(\alpha,2n)$ Gd^{149} reaction was determined by counting the samples in the 100-channel gamma pulse-height analyzer and integrating the 9-day, 150-kev peak in each plate. A function for the 29-hour activity was determined in the same way using the 229-kev peak, and its form and position are clearly those expected of an $(\alpha,4n)$ reaction. Thus, we have assigned the 29-hour activity to Gd^{147}.

The best half-life determined for Gd^{147} was measured by following the decay of the 229-kev photopeak with the 100-channel gamma analyzer. This half-life of 29 hours is consistent with half-lives of 30 hours found in resolving K x-ray and Geiger-Mueller decay curves into their Gd^{147} and Gd^{149} components.

Figure I gives the excitation functions for alpha-induced reactions on Sm^{147}. It was not possible to calculate absolute cross sections with any confidence because of lack of knowledge of the detailed decay schemes of Gd^{147} and Gd^{149} and of the half-life of Gd^{148}.

RELATIVE PHOTON AND ELECTRON INTENSITIES OF Gd^{147} AND Gd^{149} GAMMA RAYS

Figure II shows the gamma spectrum of Gd^{147}, and Figure III gives the spectrum of Gd^{149}. Since energies were accurately known from permanent-magnet

spectrograph work, complex peaks could be resolved with some confidence, and where peaks were small and indefinite, upper limits for photon intensities could be set. The values of Davisson and Evans[13] were used in correcting for absorption through lead absorbers. Curves from Kalkstein and Hollander[14] were used to obtain NaI crystal counting efficiencies. No photopeaks were seen for any of those transitions not assigned to the decay of a particular gadolinium isotope. In later runs on the 100-channel gamma analyzer the 120- and 210-kev transitions from the decay of Eu^{147} were seen. Relative electron intensities were measured with the double-focusing spectrometer and also from the permanent magnet films by use of a photodensitometer. Because of the many corrections necessary in obtaining intensities from the densitometer, more meaning is attached to the values obtained from the double-focusing spectrometer.

Table II gives the intensity values for some of the gammas of Gd^{147} and Gd^{149}. In the case of Gd^{149} the electron intensities are given relative to an intensity of 1.00 for the K-line of the 346.7-kev transition, and in the case of Gd^{147} the same thing is done using the K-line of the 396.2-kev transition. To put the relative photon intensities on the same scale as the electron intensities, multipolarities for the 149.9-kev transition of Gd^{149} and 229.5-kev transition of Gd^{147} were considered. L-subshell conversion coefficients calculated by Rose[15] and K-shell conversion coefficients calculated by Sliv[16] were used in assigning multipolarities to these transitions. Since Sliv's K-shell conversion coefficients are very similar to Rose's in the region of $Z = 63$, K/L ratios could be compared without normalization. From K/L_I ratios, L subshell ratios, and K/M_I ratios, both the 149.9- and 229.5-kev transitions appear to be magnetic dipole (or possibly M2) transitions. Electric multipolarity assignment for either transition is ruled out because of the small amount of conversion in the L_{II} subshell, and higher magnetic multipole orders were not considered because of their long lifetimes and because of the lack of conversion in the L_{III} subshell. In doing coincidence work on these isotopes neither transition was found to be delayed (i.e., both lifetimes less than a millimicrosecond). Assuming that both of these transitions were magnetic dipole, the relative photon intensities were normalized to give these transitions the proper K-shell conversion coefficients. Gammas for which photons but no conversion electrons were seen are placed in parentheses. Their energies are uncertain by several kilovolts.

Table II

Relative Intensities of Gd^{147} and Gd^{149} Gammas

Gamma energy (kev)	Line	Electron intensity from double-focussing spectrometer	Electron intensity from densitometer	Photon intensity	Total transition intensity
		Gd^{147}			
229.5				12.4	15.1
	K	1.93	1.57		
	L_I	0.56	0.29		
	M_I		0.11		
	N_I		0.06		
241					0.04
	K		0.04		
261.0					0.09
	K		0.09		
310				< 0.75	< 0.88
	K		0.13		
370				2.1	2.3
	K	0.25	0.16		
396.2				5.2	6.3
	K	1.00	1.00		
	L_I	0.14	0.31		
(517)				1.1	1.1
(570)				1.2	1.2
(635)				2.6	2.6
(770)				4.6	4.6
(900)				4.9	4.9
(1080)				1.5	1.5
(1298)				0.72	0.72
		Gd^{149}			
107				< 0.32	< 0.32
126.1				< 0.26	< 0.26
149.9				8.6	13.7
	K		4.30		
	L_I	0.55	0.46		
	M_I	0.19	0.18		
	N_I		0.06		
244				< 0.33	< 0.33
272.7				< 1.0	< 1.2
	K	0.09	0.08		
	L_I-L_{II}		0.05		
	M_I		0.06		
298.8				5.9	6.5
	K	0.49	0.44		
	L_I	0.09	0.08		
346.7				3.6	4.9
	K	1.00	1.00		
	L_I	0.23	0.20		
461				< 0.32	< 0.32
496				0.32	0.32
517				0.84	0.84
534				0.76	0.76
(650)				0.55	0.55
750				< 1.4	< 1.4
790				< 1.4	< 1.4
(937)				0.67	0.67

COINCIDENCE MEASUREMENTS

Table III gives the results of coincidence measurements made on a mixed sample of Gd^{147} and Gd^{149} by Strominger.[17] At the same time he looked for positrons and was able to set limits on the positron emission to K-capture decay ratios. The limit for positron emission by Gd^{149} was determined to be $\beta^+/K < 0.4\%$ and that for Gd^{147} to be $< 1.2\%$.

Table III

Isotope	Gate gamma energy (kev)	Energy of gammas in coincidence (kev)
Gd^{149}	150	350
		500
		760
Gd^{147}	229	390
		760
		930
Gd^{149}	630	750
Gd^{149}	750	150
		630

DISCUSSION OF RESULTS

The multipolarity of the 149.9-kev transition in Gd^{149} decay has already been discussed as being either M1 or M2. Although the M1 assignment has already been assumed in normalizing the photon intensities, the M2 possibility was considered to determine if the first assumption was correct. If the 149.9-kev gamma were an M1 transition, the 346.7-kev gamma would have a K-shell conversion coefficient of $0.16 \pm .02$ and a K/L_I ratio of $5.0 \pm .6$. These are both consistent with an M2 transition ($\alpha_k = 0.190$; $K/L_I = 5.30$). M1, E1, and E2 assignments could be ruled out because they would have K-shell conversion coefficients lower by a factor of 4 or more. If the 149.9-kev gamma were an M2 transition, the 346.7-kev gamma would have a K-shell conversion coefficient of $1.7 \pm .2$, which is at least a factor of 9 greater than the theoretical values for transitions of multipolarity less than three. It therefore seems likely that the M1 assignment to the 149.9-kev gamma and subsequent M2 assignment to the 346.7-kev gamma are correct. In addition, an assignment could be made to the 298.8-kev gamma. This transition has a

K-shell conversion coefficient of 0.08 ± .01 and a K/L_I ratio of 5.5 ± .7, both consistent with an M1 ($\alpha_K = 0.077$; $K/L_I = 5.92$). E2 was ruled out as a possible assignment because a fairly strong L_I line was seen but no L_{II} or L_{III}. If the transition were an E2, the L_{II} line would definitely be seen. M2 was ruled out because of its high α_K, E1 because of its low α_K, and higher multipole orders because of their long lifetimes. Nothing may be said about the multipole orders of the other transitions, except that of the 272.7-kev gamma. E1 may be ruled out as a multipolarity for this transition because the theoretical value of α_K for an E1 transition is 0.017 and the 272.7-kev gamma has a minimum $\alpha_K \geq 0.064 \pm .008$. No definite multipolarity assignments can be made to any of the Gd^{147} transitions.

Figure IV shows partial decay schemes for Gd^{147} and Gd^{149}. These schemes are uniquely consistent with the available energy, intensity, and coincidence data, but are far from complete. Their complexity is evidently such that, without further information about each transition, the gammas may be fitted into several different schemes. An IBM 650 computer program[18] was used to determine all cases where sums of two transition energies agree within 0.1% with another energy or sum of two other energies. Table IV tabulates the results of this comparison. The transition-energy values are reported to a greater number of significant figures in this table than they are elsewhere in the paper; although the absolute values of the energies are not known to better than 0.5%, the relative energy values are probably somewhat better.

Table IV

Transition Energy	Code Letter	Transition Energy	Code Letter
$106._6$	A	298.8	J
$119._8$	B	346.7	K
126.1	C	$460._6$	L
$132._0$	D	$496._5$	M
149.9	E	$516._7$	N
$244._3$	F	$534._2$	O
$252._7$	G	$749._5$	P
272.7	H	$789._8$	Q

Sums Found

E + K = M	D + O = E + N
F + G = M	D + P = K + O
F + H = N	E + K = F + G
G + M = P	F + P = L + O
H + N = Q	G + N = H + M
	H + O = K + L

The number of sums of two energies with one energy expected by chance is 1.4 and the number of sums of two with two expected by chance is 8.2. For this reason one may attach some statistical significance to the sums of two with one, but probably not to those of two with two.

The 496.6-kev state in the levels of Eu^{149} is of special interest because of possible isomerism, since it is depopulated by a 346.7-kev M2 transition. The theoretical mean life for this transition, using Moszkowski's[19] formula, was calculated to be $\sim 6 \times 10^{-8}$ sec. Thus, the lifetime of this state is probably measurable by delayed-coincidence techniques, but we have not had the opportunity to attempt the measurement.

No rotational structure is immediately obvious in the levels of Eu^{147} and Eu^{149}. In Coulomb excitation work Heydenburg and Temmer[20] found a definite ground-state rotational band in Eu^{153} (90 neutrons) and distorted rotational structure in Eu^{151} (88 neutrons). The present work indicates that Eu^{147} and Eu^{149} have very complex level schemes without apparent rotational band structure. Their proximity to the closed 82-neutron shell probably favors a spherical rather than spheroidal nuclear equilibrium shape. Some theoretical treatments[21,22] have correlated levels and properties of even-even nuclei in this intermediate region between the closed shells and onset of stable spheroidal deformation. Theoretical work on odd-mass nuclei of the intermediate region would be of greatest interest.

We wish to acknowledge the assistance of Dr. Jack M. Hollander in interpreting results obtained from the permanent-magnet spectrographs and of Dr. Donald Strominger in making coincidence measurements. We also wish to thank W. E. Jones and the crew of the Berkeley 60-inch cyclotron for help with bombardments and Health Chemistry for monitor service and target transportation. This work was performed under the auspices of the U.S. Atomic Energy Commission.

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FIGURE CAPTIONS

Figure I — Excitation function for alpha-induced reactions on Sm^{147}. Ordinates are in arbitrary units.

Figure II — Sodium iodide scintillation gamma spectrum of Gd^{147} (background subtracted). The 150-kev peak is the strongest photopeak of Gd^{149}, present in small abundance in the Gd^{147} sample.

Figure III — Sodium iodide scintillation gamma spectrum of Gd^{149} (background subtracted).

Figure IV — Partial decay scheme for Gd^{147} and Gd^{149}.

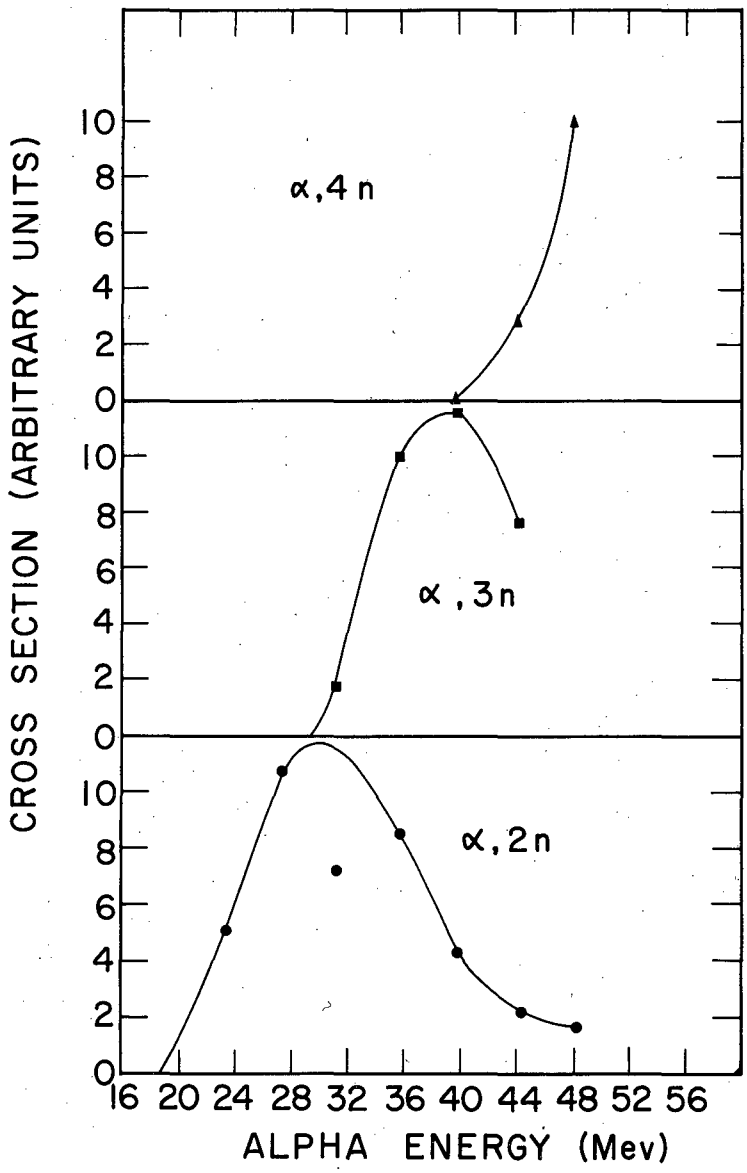
CROSS SECTION (ARBITRARY UNITS)
α,4 n
α ,3n
α,2n
0
2
4
6
8
10
16 20 24 28 32 36 40 44 48 52 56
ALPHA ENERGY (Mev)

Figure 1

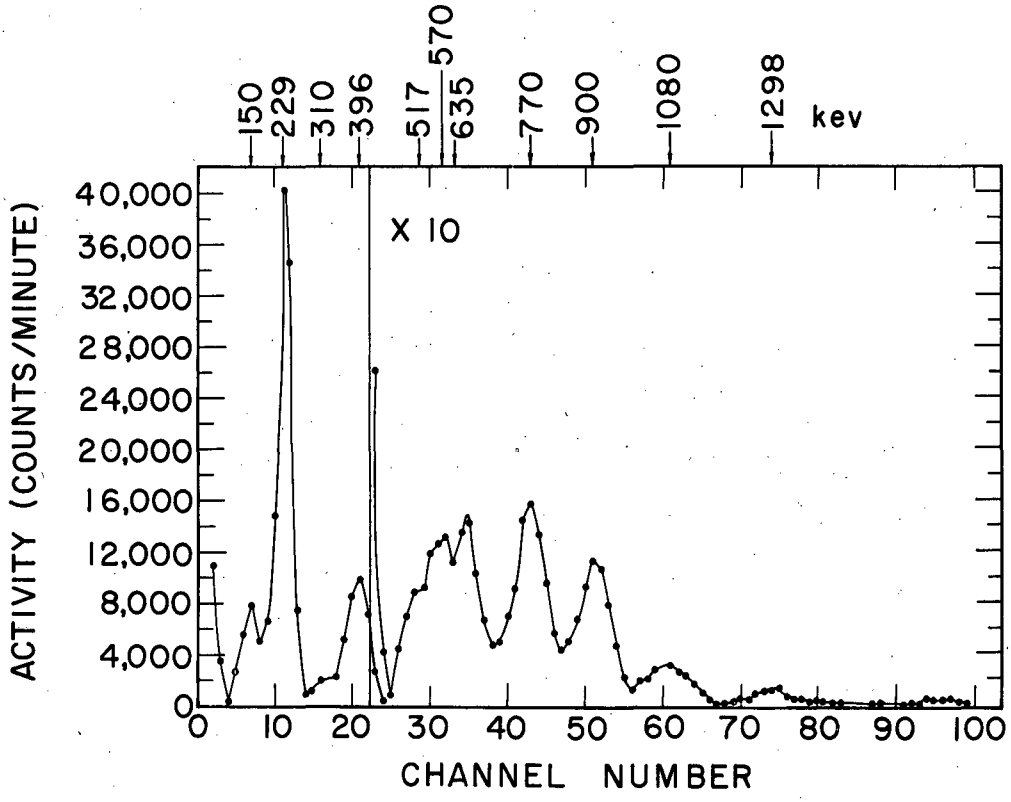

Figure 2

MU-12414

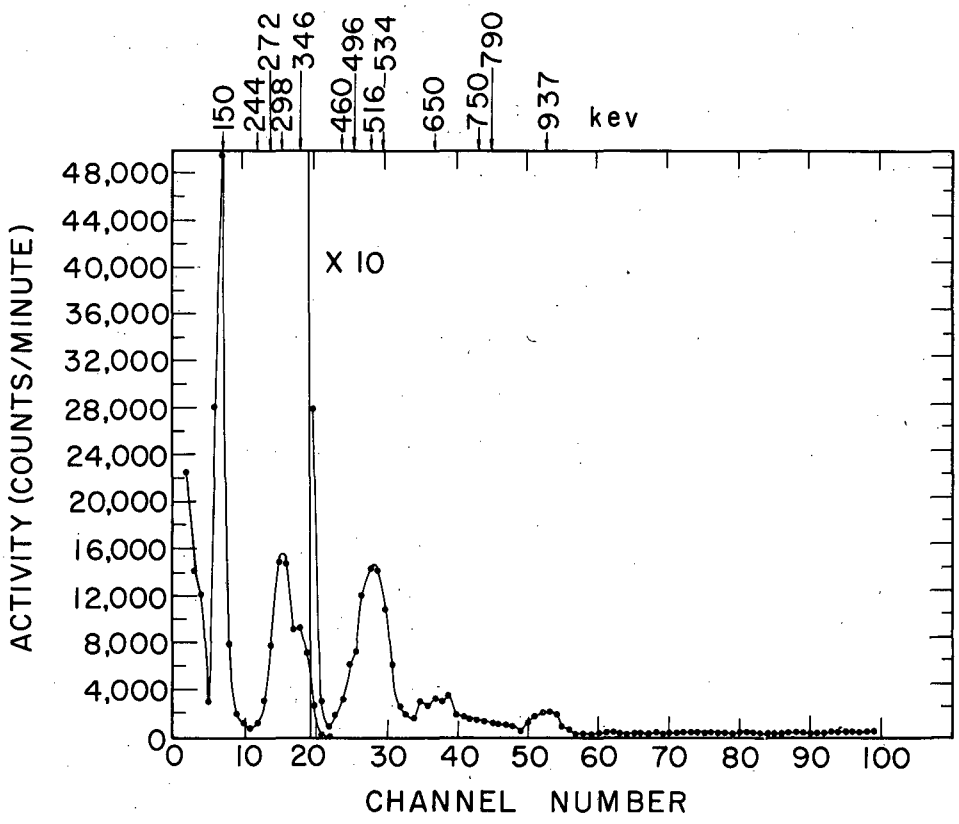

Figure 3

MU-12415

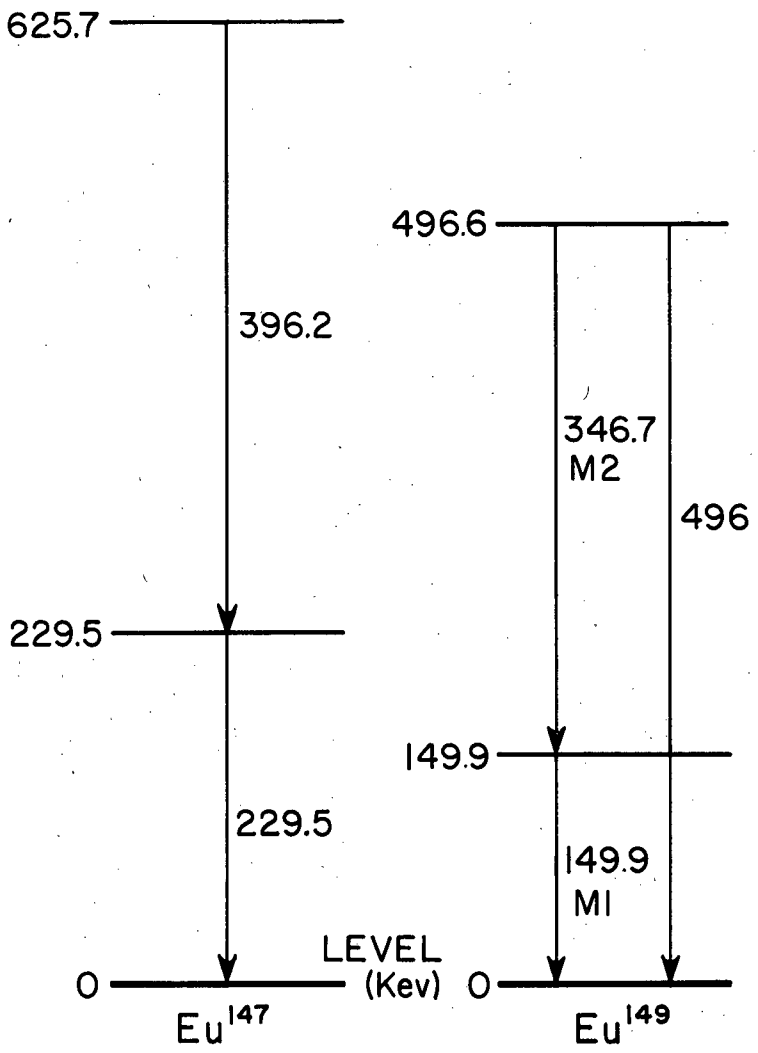

Figure 4

MU-12925