

a smaller fraction of the total energy. For the especially narrow charge 1 line this situation occurs when about 50 volts are applied to the source volume. Actually we made two careful examinations of the charge 1 peak, one at about 50 volts and one at about 90 volts. The results are plotted together in Fig. 8. Background has not been subtracted because it is instructive to leave it explicitly indicated. The working scales are indicated at the top, and through the calibration of the instrument these have been related to the recoil energy scale shown at the bottom. The two peaks are quite symmetric, and we have supposed it legitimate to use the center line as the indicator of the peak position; the same procedure was used in the calibration. The 90-volt peak centers at 9.64 ev with an uncertainty of  $\pm 0.02$  ev in the centering, and the 50-volt peak centers at 9.61 ev with an uncertainty of  $\pm 0.03$  ev. It seems acceptable to adopt  $9.63 \pm 0.02$  ev as the average, but to this we must attach an error of  $\pm 0.05$  ev as a possible calibration inaccuracy. Thus we arrive at a final value  $9.63 \pm 0.06$  ev for the energy of the A<sup>37</sup> recoils. It will be recalled that the value based upon the Cl<sup>37</sup>(*p,n*)A<sup>37</sup> threshold was expected to be  $9.65 \pm 0.05$  ev.

It is of interest to see what limit this determination places upon the rest mass of the neutrino. The momentum balance is expressed through the equation  $2ME_r = (T/c^2)(T+2\nu c^2)$ , whence it is apparent that if the neutrino rest energy  $\nu c^2$  were to amount to 1% of its kinetic energy *T*, that is, if it were to amount to 4 kev, then the expected recoil energy would be  $9.75 \pm 0.05$  instead of  $9.65 \pm 0.05$  ev, and this would be just outside

of the estimated errors so far as agreement with our determination is concerned. It is safer to state that from the evidence the neutrino rest energy is probably less than 5 to 6 kev. This limit is of course less restrictive than is the limit obtained from the shape of the high-energy end of the tritium beta spectrum, but it is based upon an independent experimental method and one that involves theoretical considerations no more complicated than the conservation of momentum. The conclusion also rests upon the assumption that the Cl<sup>37</sup>(*p,n*)A<sup>37</sup> threshold can legitimately be taken as determining the *Q*-value for that reaction.

In conclusion we may state that we have apparently encountered none of the expected difficulties that are supposed to beset the deflection of ions of low energy—that is, difficulties related to contact potentials and the charging-up of surfaces. Aside from reasonable (but not extreme) cleanliness, the only precautions that we took were to use mercury diffusion pumps to avoid internal surface films of insulating oil, and to have the ions see only stainless steel throughout their flight. Perhaps the saving features were that the apertures were large and the current densities extremely small. We expect to use the spectrometer in the measurement of continuous recoil spectra accompanying the beta decay of rare gases, but here an additional stage of differential pumping will have to be added because one then will forego the concentration of the recoil intensity into lines. Meanwhile, the method has proved itself under rigorous conditions in the detailed investigation of the very low-energy A<sup>37</sup> line spectrum.

### Radioactive Decay of the Isomers of Americium-242\*

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 (Received May 23, 1955)

The decay characteristics of the two isomers of Am<sup>242</sup> have been investigated. The beta decay of the 16-hour Am<sup>242m</sup> has a branching ratio of  $51 \pm 5\%$  to the first-excited state (42.3 kev) of Cm<sup>242</sup> with the remainder of the decay going to the ground state. The electron capture decay of Am<sup>242m</sup> has a branching ratio of approximately 60% to first-excited state (44.8 kev) of Pu<sup>242</sup>. An upper limit of 6% has been set for the fraction of Am<sup>242m</sup> decay via isomeric transition. The beta decay of the 100-year Am<sup>242</sup> has a branching ratio of  $45 \pm 5\%$  to the first-excited state of Cm<sup>242</sup>, with the remainder of the decay going to the ground state. The beta-spectrum end points for Am<sup>242m</sup> and Am<sup>242</sup> have been measured to be  $620 \pm 10$  kev and  $585 \pm 10$  kev, respectively. A decay scheme for the two isomers has been proposed. *Logft* values have been calculated for beta and electron capture decay of the isomers and are discussed in conjunction with spin and parity assignments.

#### I. INTRODUCTION

THE two isomers, 16-hour Am<sup>242m</sup> and long-lived (approximately 100 years) Am<sup>242</sup>, were first observed as neutron-capture products of Am<sup>241</sup>.<sup>1</sup> The

decay characteristics of these isomers, particularly Am<sup>242m</sup>, have been studied by O'Kelley *et al.*<sup>2</sup> From the experimental data obtained by the authors, it is possible to formulate decay schemes for these isomers which differ in certain respects from those reported in

\* This work was performed under the auspices of the U. S. Atomic Energy Commission. The contents of this report have been presented in somewhat greater detail in University of California Radiation Laboratory Unclassified Reports UCRL-2325, UCRL-2528, and UCRL-2537 (unpublished).

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<sup>1</sup> Seaborg, James, and Morgan, *The Transuranium Elements*:

*Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV, p. 1529; Seaborg, James, and Ghiorso, NNES, p. 1554; W. M. Manning and L. B. Asprey, NNES, p. 1595.

<sup>2</sup> O'Kelley, Barton, Crane, and Perlman, *Phys. Rev.* **80**, 293 (1950).

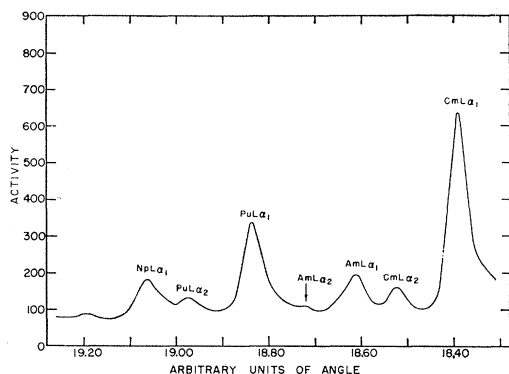


FIG. 1.  $L\alpha$  x-ray spectrum of a sample containing  $\text{Am}^{241}$  and  $\text{Am}^{242m}$ .

previous work. The fraction of  $\text{Am}^{242m}$  decay which proceeds via the isomeric transition was found to be much smaller than was thought previously. Also, the energies of the levels of  $\text{Pu}^{242}$  and  $\text{Cm}^{242}$  populated by the decay of  $\text{Am}^{242m}$  have been determined from a re-interpretation of the conversion electron spectrum. The results of Church<sup>3</sup> on the decay of  $\text{Am}^{242m}$  essentially agree with the data reported here.

## II. EXPERIMENTAL METHODS AND RESULTS

The measurements described were performed on quantities of  $\text{Am}^{242m}$  and  $\text{Am}^{242}$  produced in three separate neutron irradiations of purified  $\text{Am}^{241}$ . The decay characteristics of  $\text{Am}^{242m}$  were studied using americium from two irradiations of one day and five days in duration.<sup>4</sup> A separate sample of americium which had been subjected to a much greater total neutron irradiation was used in the study of the long-lived  $\text{Am}^{242}$ . Mass analysis of this sample revealed the following isotopic abundances:  $\text{Am}^{241}$ —90.4%,  $\text{Am}^{242}$ —1.1%, and  $\text{Am}^{243}$ —8.5%. The americium was purified after irradiation using a combination of hydroxide and fluoride precipitations, ion exchange columns, and oxidation to the hexapositive oxidation state, techniques which have been described previously.<sup>5</sup> Since the  $\text{Am}^{242}$  isomers were produced by neutron irradiation,  $\text{Am}^{241}$  constituted a large fraction of the mass of each sample. The radiations of  $\text{Am}^{242m}$  were identified by their 16-hour decay, while the radiations of long-lived  $\text{Am}^{242}$  were resolved in a mixture of  $\text{Am}^{241}$  and  $\text{Am}^{242}$  by eliminating those which also appeared in a pure sample of  $\text{Am}^{241}$ .

The electromagnetic radiation of  $\text{Am}^{242m}$  in the region of 13–60 keV was examined with a bent-crystal spectrometer of the Cauchois type.<sup>6</sup> A portion of this

<sup>3</sup> E. L. Church (Argonne National Laboratory, unpublished report, 1953).

<sup>4</sup> The irradiations were performed in the Materials Testing Reactor at the National Reactor Testing Station, Idaho Falls, Idaho.

<sup>5</sup> Thompson, Ghiorso, and Seaborg, *Phys. Rev.* **80**, 781 (1950); Asprey, Stephanou, and Penneman, *J. Am. Chem. Soc.* **72**, 1425 (1950).

<sup>6</sup> Barton, Robinson, and Perlman, *Phys. Rev.* **81**, 208 (1951).

spectrum showing the  $L\alpha$  lines is shown in Fig. 1. The  $L$  x-rays of neptunium observed in the spectrum arise from the conversion of gamma rays from the alpha decay of  $\text{Am}^{241}$ . The data on the  $L$  x-rays associated with the decay of  $\text{Am}^{242m}$  are listed in Table I. In addition to the bent-crystal spectrometer study, a sodium-iodide-crystal spectrometer was employed to measure the intensity of the Pu  $K$  x-ray. Using fluorescence yields based on Kinsey's estimates,<sup>7</sup> the relative  $L$  shell vacancies have been calculated for curium and plutonium:  $\text{Cm } L_2:L_3=250:110$  and  $\text{Pu } K:L_2:L_3=40:70:30$ . These figures were based upon the observed intensities and were corrected for the abundance of  $L$  lines not observed using data on  $\text{Am}^{241}$ .<sup>8</sup> The  $L_1$  quantum yield has been omitted because of uncertainties in line assignments and low abundance. The fluorescence yield for the plutonium  $K$  shell is estimated as 98%.<sup>9</sup> A correction has also been applied for the  $L$  vacancies in plutonium due to emission of  $K$  x-rays. No corrections have been made to the above intensity ratios for non-radiative transitions of the Coster-Krönig type because of the uncertainty in the magnitude of these effects. These  $L$  x-ray intensities lead to a ratio of curium to plutonium  $L$  conversion of 3.6 assuming only  $K$  and  $L_1$  electron capture.

There is considerable question as to the origin of the americium  $L$  x-rays. It was observed that these lines decayed to some extent and then remained constant with time. The americium  $L$  x-rays which did not

TABLE I. X-rays following the decay of  $\text{Am}^{242m}$ .

Line	Transition	Observed energy (keV)	Siegbahn's extrapolated energy (keV)	Corrected relative intensity
Cm $L\alpha_2$	$L_3-M_4$	$14.75 \pm 0.03$	14.74	4
Cm $L\alpha_1$	$L_3-M_5$	$14.97 \pm 0.03$	14.96	27
Cm $L\beta_2$	$L_3-N_5$	$18.09 \pm 0.02$	18.10	16
Cm $L\beta_1$	$L_2-M_4$	$19.47 \pm 0.02$	19.38	100
Cm $L\gamma_1$	$L_2-N_4$	$22.79 \pm 0.04$	22.63	40
Cm $L\gamma_3$	$L_1-N_3$	$23.30 \pm 0.06$	23.25	6
Cm $L\gamma_6$	$L_2-O_4$	$23.62 \pm 0.12$	23.46	12
Am $L\alpha_2$	$L_3-M_4$	$14.44 \pm 0.06$	14.41	1
Am $L\alpha_1$	$L_3-M_5$	$14.61 \pm 0.03$	14.61	4
Am $L\beta_1$	$L_2-M_4$	$18.89 \pm 0.02$	18.80	6
Pu $L\alpha_2$	$L_3-M_4$	$14.08 \pm 0.03$	14.08	2
Pu $L\alpha_1$	$L_3-M_5$	$14.28 \pm 0.03$	14.28	10
Pu $L\beta_2$	$L_3-N_5$	$17.29 \pm 0.03$	17.25	9
Pu $L\beta_1$	$L_2-M_4$	$18.33 \pm 0.03$	18.27	27
Pu $L\beta_3$	$L_1-M_3$	$18.62 \pm 0.04$	18.52	7
Pu $L\gamma_1$	$L_2-N_4$	$21.46 \pm 0.04$	21.38	16
Pu $L\gamma_3$	$L_1-N_3$	$22.06 \pm 0.10$	21.97	7
Pu $L\gamma_6$	$L_2-O_4$	$22.24 \pm 0.10$	22.13	7
Pu $K$ x-rays		102		37

<sup>7</sup> B. B. Kinsey, *Can. J. Research* **26A**, 404 (1948).

<sup>8</sup> Jaffe, Passell, Browne, and Perlman, *Phys. Rev.* **97**, 142 (1955).

<sup>9</sup> E. H. S. Burhop, *The Auger Effect and Other Radiationless Transitions* (Cambridge University Press, Cambridge, 1952), p. 48.

decay with a characteristic 16-hour half-life are thought to be due to self-excitation of the bulk  $\text{Am}^{241}$  in the sample, and a correction for this effect has been applied to the intensities. In addition, the presence of x-rays with the  $\text{Am}^{242m}$  decay capable of exciting the americium  $L_2$  and  $L_3$  shells indicates the observed relative abundances are merely upper limits. From the intensity of the americium  $L$  x-rays and on the basis of  $L$  conversion alone, an upper limit of 6% may be set for a predominantly  $L$ -converted isomeric transition. Although no evidence has been found for the presence of radiations or conversion electrons of an isomeric transition of energy greater than about 30 keV, there is a possibility that the isomeric transition is either not energetic enough to convert in the  $L$  shell of americium or produces extremely low-energy  $L$  conversion electrons. In such a case the upper limit of 6% for the isomeric transition does not hold.

The conversion electron and beta spectra of these isomers were studied by use of a double-focusing  $\pi\sqrt{2}$  magnetic beta-ray spectrometer. The energies and relative intensities of the electron lines of  $\text{Am}^{242m}$  are listed in Table II. The absolute uncertainty on the electron energies is  $\pm 1.5$  keV. The present interpretation of these conversion electron lines indicates they arise from the  $L$ -shell conversion of two gamma rays. The two most abundant lines were assigned to the  $L$  conversion of a 41-keV gamma ray in  $\text{Cm}^{242}$ . The 43-keV gamma ray is thought to be present in the electron capture branch of the decay to  $\text{Pu}^{242}$  giving rise in part to the observed  $L$  x-rays of plutonium. The conversion electron intensities yield a ratio of curium to plutonium  $L$  shell vacancies of 2.5. It should be noted that these intensities are subject to uncertainties in resolution, scattered electron background, and an unknown contribution from  $L$  Auger electrons. The  $L_2/L_3$  conversion ratios are  $1.4 \pm 0.4$  for each gamma ray. Since  $E1$  and most magnetic radiations are expected to exhibit marked  $L_1$  conversion,<sup>10</sup> the radiation is most likely electric quadrupole or higher multipole electric radiation. The assignment of  $E2$  character to both transitions is consistent with the theoretical calculations of Gellman *et al.*<sup>11</sup> and the regularities noted among first excited states of even-even nuclei.<sup>12</sup>

Two gamma rays of 42.3- and 44.8-keV energy were barely discernible with the bent crystal spectrometer. Their energy difference compares favorably with a 2.3-keV difference from conversion electron data. Experiments performed by Church give energies of  $42.2 \pm 0.3$  and  $44.6 \pm 0.3$  keV, respectively, for the gamma rays following  $\text{Am}^{242m}$  beta and electron capture decay.<sup>3</sup> These energies agree well with those from bent crystal spectrometer measurements and are considered to be the best values. The absence of conversion electron lines

TABLE II. Electron energies and intensities for  $\text{Am}^{242m}$ .

Gamma energy (keV)	Electron energy (keV)	Conversion shell	Intensities (arbitrary units)
41.0	17.3	Cm $L_{II}$	520
	22.0	Cm $L_{III}$	380
	35.5	Cm $M_{II}$	380
	36.5	Cm $M_{III}$ or $M_{IV}$	
	39.9	Cm $N$ (Pu $M?$ )	
43.3	20.9	Pu $L_{II}$	210
	25.5	Pu $L_{III}$	150
Cm Auger electrons	$\sim 16.5$	$L:MM, L:MN, L:NN$	500

of any other gamma ray is consistent with the low intensity of the isomeric transition but does not preclude an isomeric transition of extremely low energy.

The beta spectrum of each isomer was determined with separate samples. Results agreed with previous determinations,<sup>2</sup> the beta end point of  $\text{Am}^{242m}$  occurring at  $620 \pm 10$  keV while that of  $\text{Am}^{242}$  was a  $585 \pm 10$  keV. No conversion electron lines were observed in the beta spectrum of the long-lived  $\text{Am}^{242}$  sample which could not reasonably be ascribed to the 60 and 43 keV transitions in the decay of  $\text{Am}^{241}$ . Beta- $L$  x-ray coincidence measurements were performed on both isomers using an anthracene crystal for detecting beta particles and a thallium-activated sodium iodide crystal with a beryllium window for observing  $L$  x-rays. The  $L$  x-rays due to the alpha decay of  $\text{Cm}^{242}$  which has a branching ratio of 26.3% to the first-excited level of  $\text{Pu}^{238}$  were used as a standard in determining the branching ratios of the  $\text{Am}^{242}$  isomer beta decay. A comparison of an  $\text{Am}^{242m}$  sample with the  $\text{Cm}^{242}$  standard gave a branching ratio of  $51 \pm 5\%$  to the first-excited state of  $\text{Cm}^{242}$  in the beta decay of  $\text{Am}^{242m}$ , after suitable corrections were made for differences in fluorescence yield and absorption of  $L$  x-rays. From a similar measurement on  $\text{Am}^{242}$ , a branching ratio of  $45 \pm 10\%$  was calculated. The remainder of the beta transitions, in each case, proceed to the ground state of  $\text{Cm}^{242}$ . Thus the beta spectrum of each isomer should consist of two components differing by approximately 43 keV in energy. These components were not resolved experimentally. If it is assumed that the measured beta particle energies are the respective ground-state transitions, the separation between the two  $\text{Am}^{242}$  isomers is approximately 35 keV. However, in view of the uncertainties in the beta spectrum end points and the inability to resolve each spectrum into its components, the energy spacing between the  $\text{Am}^{242}$  isomers is rather uncertain.

The branching ratio  $\beta^-/\text{EC} = 4.2$  for  $\text{Am}^{242m}$  has been reported from an analysis of products from a long neutron irradiation of  $\text{Am}^{241}$ .<sup>13</sup> The ratio of curium to plutonium  $L$  shell vacancies has been determined by two experimental methods,  $L$  x-ray (3.6) and conversion

<sup>13</sup> G. H. Higgins and S. G. Thompson, February, 1953 (unpublished).

<sup>10</sup> E. L. Church and J. E. Monahan, Phys. Rev. **98**, 718 (1955).

<sup>11</sup> Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).

<sup>12</sup> G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953); F. Asaro and I. Perlman, Phys. Rev. **87**, 393 (1952).

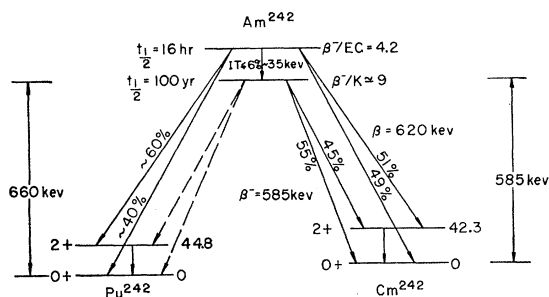


FIG. 2. Decay scheme for the isomers of  $\text{Am}^{242}$ .

electron (2.5) intensities. These data may be used in conjunction with the experimentally known ratio of beta decay of  $\text{Am}^{242m}$  to the ground and first-excited states of  $\text{Cm}^{242}$  and the experimental  $\beta^-$  to electron capture branching ratio to calculate an electron capture branching ratio to the ground and first-excited states of  $\text{Pu}^{242}$ . Using  $L$  x-ray intensity data, the electron capture branching of  $\text{Am}^{242m}$  to the first-excited state of  $\text{Pu}^{242}$  is 60%, while if the conversion electron intensity data are used, the branching decay to the first excited level of  $\text{Pu}^{242}$  is 87%. The lower value, 60%, is thought to be more accurate in that the electron capture branch of the  $\text{Am}^{242m}$  decay is expected to be very similar to the  $\beta^-$  branch since the energy levels of the daughter nuclei,  $\text{Pu}^{242}$  and  $\text{Cm}^{242}$ , are similar and the energies for beta decay and electron capture (closed decay cycles)<sup>14</sup> are nearly equal.

The  $K$  electron capture branching of  $\text{Am}^{242}$  was detected with the scintillation spectrometer by observing plutonium  $K$  x-ray radiations in a sample of americium 241, 242, and 243 from which the  $\text{Np}^{239}$  had been removed chemically. (The product of  $\text{Am}^{243}$  alpha decay,  $\text{Np}^{239}$ , has prominent plutonium  $K$  x-rays associated with it.) It was also necessary to employ a silver absorber to decrease the intensity of the 60-keV gamma ray of  $\text{Am}^{241}$ . Making suitable correction for absorption, escape peak, and the presence of a 75-keV gamma ray of  $\text{Am}^{243}$ , the ratio of  $K$  x-rays to 60-keV gamma was calculated. Combining this information with the known mass composition of the sample, a ratio of  $\beta^-$  to  $K$  electron capture of approximately 9 was calculated for  $\text{Am}^{242}$ .

### III. DECAY SCHEME

The decay scheme of the  $\text{Am}^{242}$  isomers is shown in Fig. 2. The energies of the first-excited levels of  $\text{Pu}^{242}$  and  $\text{Cm}^{242}$  compare favorably with those of other even-even nuclei of plutonium and curium.<sup>15</sup> In addition,

<sup>14</sup> Glass, Thompson, and Seaborg, *J. Inorg. Chem. Nuc. Chem.* **1**, 3 (1955).

<sup>15</sup> Hollander, Perlman, and Seaborg, *Revs. Modern Phys.* **25**, 469 (1953).

the complex alpha structure of  $\text{Cf}^{246}$  shows an energy spacing of 42.7 keV for the first-excited level of  $\text{Cm}^{242}$  which is in excellent agreement with the value determined here.<sup>16</sup> If one uses the measured beta energy and branching ratio, the  $\log ft$  for both beta transitions of  $\text{Am}^{242m}$  is 6.9. Similarly, the  $\log ft$  for electron capture to both levels of  $\text{Pu}^{242}$  is 7.3 as calculated from equations for allowed  $f^{17}$  using energies derived from closed decay cycle calculations.<sup>14</sup> These  $\log ft$  values indicate first forbidden transitions,  $\Delta I=0$ , 1 yes, for both beta and electron capture decay. The long-lived  $\text{Am}^{242}$  has a  $\log ft$  of 11.6 for beta decay to both levels of  $\text{Cm}^{242}$ . Although the percentage of electron capture decay to the ground and first-excited state of  $\text{Pu}^{242}$  is unknown for this isomer, a  $\log ft$  of 12.3 for  $K$  electron capture may be calculated if one assumes approximately equal population of these two levels. These rather large  $\log ft$  values for  $\text{Am}^{242}$  may indicate second forbidden transitions.

The spins of the two  $\text{Am}^{242}$  isomers are still undetermined. Since  $\text{Am}^{242m}$  decays to both the ground and first-excited states of its daughters with approximately equal probability as evidenced by the  $ft$  values, a spin of unity with odd parity would be consistent with the first forbidden beta and electron capture decay. There are fewer data available on the decay of the long-lived  $\text{Am}^{242}$ . Since the beta decay of this isomer also apparently populates the ground and first-excited states of  $\text{Cm}^{242}$  with equal probability, a relatively low spin is indicated. The absence of an appreciable isomeric transition may be explained either by a very long half-life due to a large spin difference between isomers or by a very small energy difference between isomers, which would make the isomeric transition difficult to detect. At present the problem of spin and parity assignments must be left unresolved. Perhaps the most useful information for solving this problem would be a further study of the decay characteristics of the long-lived  $\text{Am}^{242}$ .

### ACKNOWLEDGMENTS

It is a pleasure to acknowledge the continued interest and helpful suggestions of Professor G. T. Seaborg and Professor I. Perlman. We wish to thank Dr. G. H. Higgins for his assistance in the early phases of the chemical separations. Discussions of this problem with Dr. J. O. Rasmussen, Jr., have been most helpful. We are also grateful to Dr. W. B. Lewis and the Materials Testing Reactor Staff for aid in the irradiation of the sample.

<sup>16</sup> Hummel, Stephens, Asaro, Chatham-Strode, and Perlman, *Phys. Rev.* **98**, 22 (1955).

<sup>17</sup> J. K. Major and L. C. Biedenharn, *Revs. Modern Phys.* **94**, 779 (1954).