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# The Alpha Spectra of $Cm^{242}$, $Cm^{243}$, and $Cm^{244}$

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The alpha and gamma spectra of $Cm^{242}$, $Cm^{243}$, and $Cm^{244}$ have been studied with an alpha-particle spectrograph and gamma-ray scintillation counters. $Cm^{242}$ has alpha groups of 6.110 (73.7 percent), 6.066 (26.3 percent), and 5.964 Mev (0.035 percent) and gamma rays of 44 (0.041 percent), 100 (0.006 percent), and 157 kev (0.0027 percent). $Cm^{243}$ has alpha groups of 5.985 (6 percent), 5.777 (81 percent), and 5.732 Mev (13 percent) and gamma rays of 104, 226, and 278 kev in coincidence with the 5.777-Mev alpha group. $Cm^{244}$ has alpha groups of 5.798 (75 percent) and 5.755 Mev (25 percent). The spectra are discussed relative to alpha-decay theory and corresponding excited states reached by $\beta^-$ decay processes.

## I. INTRODUCTION

THE examination of alpha spectra has been continued using the 75-cm radius of curvature, 60° sector electromagnetic analyzer described previously.[1,2] Certain improvements in the techniques of sample preparation and in the calibration and operation of the spectrograph have resulted in greater refinement of the measurements. As an example of the results, a new rare alpha group in the decay of $Cm^{242}$ has been found, whereas previously[2] only two abundant groups were discernible. Two prominent alpha groups separated by a characteristic energy have been encountered repeatedly among the even-even nuclides,[3] and the implication of a "third alpha group" will be discussed later.

The major objective of the present study was to determine the spectra of curium isotopes of higher mass number than the familiar $Cm^{242}$. Since these isotopes are produced from $Cm^{242}$ by successive neutron capture or through americium isotopes by a similar mechanism, they are not obtainable in isotopically pure state, and the spectra must be resolved from mixtures. It will be seen that the energy spread of alpha groups from $Cm^{243}$ overlaps the $Cm^{242}$ and $Cm^{244}$ groups and an instrument of high resolution is necessary for their separation. This situation is similar to that encountered[4] with the alpha spectra of $Pu^{239}$ and $Pu^{240}$ in which it was found that two of the $Pu^{239}$ alpha groups fell between the two groups of $Pu^{240}$. Assignment of the several groups to particular isotopes was made by measuring samples of altered isotopic composition.

The present study also gives further information for the developing systematics and theory of complex alpha spectra. It will be seen that $Cm^{244}$ has a spectrum virtually identical with other even-even alpha emitters of the heaviest elements, and that $Cm^{243}$ displays the pattern of alpha emitters with odd nucleons. The hindrance of decay directly to the ground state for odd nuclear cases is again in evidence; in fact, none of the alpha groups seen so far from $Cm^{243}$ represents the ground-state transition.

## II. METHODS AND INSTRUMENT CALIBRATION

All samples which were used as sources in the spectrograph were prepared by vacuum sublimation. After chemical purification each sample of curium present in solution as the chloride was evaporated to dryness on a tungsten ribbon. Under vacuum, current was passed through the tungsten ribbon and the curium sublimed onto a 2-mil thick platinum plate masked to a band 1 in.×0.12 in. When placed in the spectrograph, the sample was made to approximate a line source by placing before it a stainless steel plate with a defining slit 1 in.×0.018 in. or 1 in.×0.005 in.

The alpha particles after magnetic analysis were intercepted on a photographic plate placed 30° to the direction of the beam and the tracks counted with a 450 power binocular microscope. The alpha spectrum was reproduced by plotting the numbers of tracks found in each $\frac{1}{4}$-mm scan of the plate.

One of the difficulties in the use of this spectrograph as a precision instrument is that the optics are not rigidly defined. The nominal radius of curvature of the magnet (75 cm) is only approximate, and the source and detector must be aligned for optimum focusing. It has been the practice to determine alpha energies by comparing with alpha emitters of known energy and by using these to correct the calculated dispersion of the instrument. The dispersion of the instrument should be given by the expression:

$$\text{dispersion}=\frac{E}{2r_0}=\frac{B^2r_0^2}{2(144)^2r_0}=\frac{B^2r_0}{2(144)^2},$$

where $B$ is the magnetic field in gauss, $r_0$ is the normal radius of curvature of the magnet in centimeters, and $E$ is the energy of the alpha particle in electron volts. It has been found upon use of the nominal radius, 75 cm, for $r_0$ that the calculated dispersion is some 6 percent lower than the measured dispersion.

Figures 1a and 1b illustrate data used to obtain the correction applied to the calculated dispersion and to show that the dispersion is essentially constant irrespective of the position on the 22-cm long plate upon which the alpha particles are focused. The circles of Fig. 1a, showing the energy difference between the two principal

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[1] F. L. Reynolds, Rev. Sci. Instr. 22, 749 (1951).
[2] Asaro, Reynolds, and Perlman, Phys. Rev. 87, 277 (1952).
[3] F. Asaro and I. Perlman, Phys. Rev. 87, 393 (1952).
[4] F. Asaro and I. Perlman, Phys. Rev. 88, 828 (1952).

alpha groups of $Cm^{242}$, were obtained on a single photographic plate by using six different magnetic field settings and employing the above expression for the dispersion with a +5.9 percent correction. The other symbols indicate other sets of data. It is seen that within the limits of determining the peak position (about one field of view in the microscope corresponding to 1-kev energy) the energy separation is constant over the part of the plate examined.

Figure 1b shows the method used for obtaining the correction term for calculating the dispersion. Sets of alpha emitters, each with two groups of known energy difference, were employed to make a series of exposures, and for each the empirical correction term for the dispersion formula was found. The alpha groups used were Rn-RaA, Rn-Po, and the two groups of $Ra^{226}$ for which the respective energy differences 512 kev, 188 kev and 183 kev were assumed. The average value for the correction term was 5.9 percent. As indicated there was a rather large uncertainty in each measurement which is partly the result of an unsatisfactory method of locating peak position no longer employed.

For present purposes the correction term and its limits of error will be taken to be 5.9 (±2) percent. This allows liberal limits for the uncertainty in instrument calibration.

The method used to determine peak position alluded to above was to extrapolate the high-energy edge of the peak to the origin. It was later noted that peaks registering on different parts of the plate had different widths. A better comparison would result from the use of the position of maximum peak height, and this convention is now used.

The disparity in peak widths at different positions of the plate was partly due to misalignment of source and detector, but part is inherent in the focusing properties of the magnet. Some improvement resulted from partial disassembly and realignment of the source and receiver assemblies. In addition, a proton resonance fluxmeter has been installed in the magnet gap so that the magnetic field can be measured directly for each run. Previously a series of flux measurements had been made as a function of magnet coil current, and this relationship had been assumed to remain constant. Data shown in Figs. 3 and 4b were obtained before source realignment while the more constant peak width following the changes is reflected in Figs. 4a, c, and d. The data shown in Fig. 1a were taken after the instrument improvements were made.

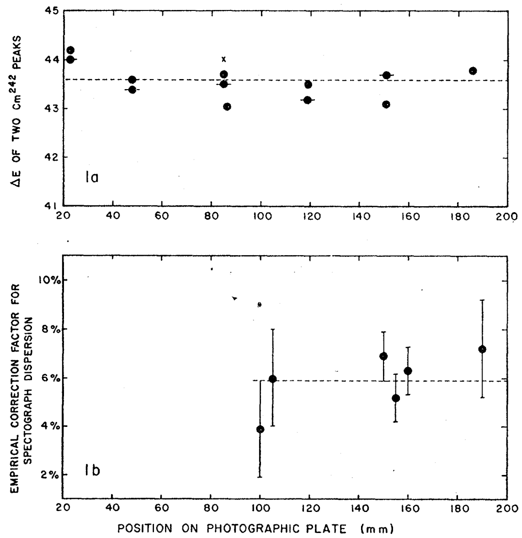
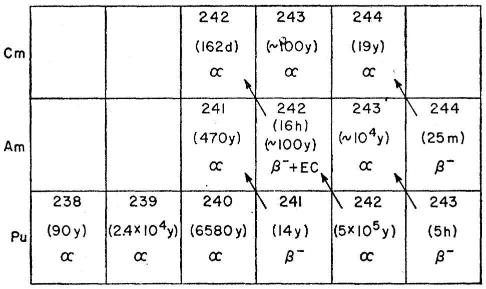


FIG. 1. a. Data illustrating uniformity of dispersion of spectrograph for focusing at different positions on photographic plate. b. Determination of the empirical correction for the dispersion formula.



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|---|---|---|---|---|---|---|
| Cm | | | 242<br>(162d)<br>α | 243<br>(~100y)<br>α | 244<br>(19y)<br>α | |
| Am | | | 241<br>(470y)<br>α | 242<br>(16h)<br>(~100y)<br>$\beta^-$+EC | 243<br>(~$10^4$y)<br>α | 244<br>(25m)<br>$\beta^-$ |
| Pu | 238<br>(90y)<br>α | 239<br>(2.4×$10^4$y)<br>α | 240<br>(6580y)<br>α | 241<br>(14y)<br>$\beta^-$ | 242<br>(5×$10^5$y)<br>α | 243<br>(5h)<br>$\beta^-$ |

FIG. 2. Segment of isotope chart showing beta-decay relationships of plutonium, americium, and curium isotopes.

### III. RESULTS

As already mentioned, the three curium isotopes under consideration could not be produced individually isotopically pure. The assignments of the alpha groups among the several curium isotopes were made principally by comparing samples with different isotopic ratios. Before discussing these, mention will first be made of the redetermination of the energy difference and relative abundances of the two principal alpha groups of $Cm^{242}$ previously reported[2] as differing in energy by 45.7 kev and having abundances of 73 percent and 27 percent. The improvements made in such determinations, such as a better knowledge of the magnetic field strength, have resulted in some revisions.

### Principal Alpha Groups of $Cm^{242}$

The difference in energy of the two groups taken as the average of the 13 values shown in Fig. 1a is 43.6 (±0.6) kev. The limits of error shown include the spread of all values. If we add to this the possible 2 percent error in instrument calibration a conservative limit of error would be ±1.4 kev. If we take the energy of the ground-state alpha group ($\alpha_0$) to be 6.110 Mev,[2] that for the other prominent group ($\alpha_{44}$) is 6.066 Mev.

The abundance of this low-energy group has been redetermined as 26.3 (±0.5) percent. Attention should be called to the fact that the difference in alpha-group energies is not quite the energy of the corresponding gamma-ray transition because of the difference in recoil energy given to the residual nuclei by the two

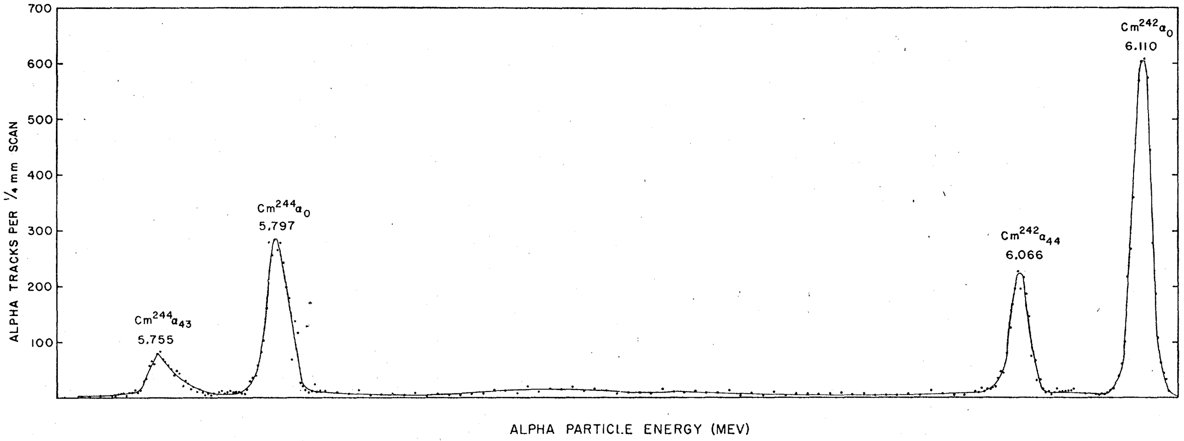

FIG. 3. Spectrum showing principal groups of $Cm^{242}$ and $Cm^{244}$ with $Cm^{243}$ absent.

groups. The energy difference between the ground state and the first excited state of $Pu^{238}$ is accordingly 44.3 kev.

### Principal Alpha Groups of $Cm^{244}$

In order better to visualize the origin of the different isotopic compositions of curium to be discussed, reference is made to a segment of an isotope chart shown as Fig. 2. References to the data used here as well as other decay data to be mentioned will be found in a new edition of the "Table of Isotopes."[5] The first sample to be mentioned was composed principally of $Cm^{242}$ and $Cm^{244}$. For its preparation, a sample of plutonium having a high concentration of $Pu^{242}$ was available, and its preparation, which is similar to one already described,[6] will be mentioned first. A sample of $Am^{241}$ was irradiated with neutrons to form 16-hour $Am^{242}$ which by its electron capture branching gives $Pu^{242}$ and through $\beta^-$ decay forms $Cm^{242}$. The preponderance of $\beta^-$ branching of 16-hour $Am^{242}$ and the relatively short alpha-decay half-life of $Cm^{242}$ conspire to produce more $Pu^{238}$ than $Pu^{242}$ if the duration of irradiation is the order of the half-life of $Cm^{242}$, or longer. With long irradiation $Pu^{238}$ undergoes further transmutation to give small quantities of higher isotopes of plutonium. The pertinent information for present considerations is that the plutonium sample so prepared contained about 25 percent $Pu^{242}$ and 0.1 percent $Pu^{241}$ by weight.

The chemically purified plutonium fraction was further irradiated, and from this the curium fraction contained roughly equal quantities by radioactivity of $Cm^{242}$ and $Cm^{244}$. The appearance of both curium isotopes is approximately second order with respect to irradiation time: $Cm^{242}$ by the slow $\beta^-$ decay of $Pu^{241}$ followed by neutron capture by $Am^{241}$, and $Cm^{244}$ from neutron capture by $Pu^{242}$ and $Am^{243}$, successively. The pertinent neutron capture cross sections which explain these results will be found in a summary by Huizenga, Manning, and Seaborg.[7]

The alpha spectrum of this mixture of $Cm^{242}$ and $Cm^{244}$ is shown in Fig. 3. The limitation in sample size ($10^6$ alpha disintegrations per minute) prevented observation of possible rare alpha groups, but the identification of the two principal alpha groups of $Cm^{244}$ relative to those of $Cm^{242}$ was unambiguous by virtue of the method of making the curium isotopes. The energies of these alpha groups were found to be 5.798 and 5.755 Mev. There were small amounts of $Am^{241}$ and $Pu^{238}$ in the sample, but their alpha groups fall at energies well below those of the new $Cm^{244}$. The data shown in Fig. 3 were obtained from a 42-hour exposure; the sample was prepared by subliming the chloride from a tungsten filament onto a 1-in.×$\frac{1}{8}$-in. band on platinum, and this band was collimated to approach a line source by a 1-in.×0.018-in. slit in a stainless steel plate.

### Low-Energy Alpha Groups of $Cm^{242}$

In an earlier study it was shown[2] that at energies lower than the two principal groups (6.110 and 6.066 Mev) there is no alpha group in abundance greater than about 0.1 percent. With the improvements made in the instrument, a weak group assigned to $Cm^{242}$ has appeared at 5.964 Mev. This group appears in all of the spectra of Fig. 4 and in another taken but not shown. (The origins of the several samples having the spectra shown in Fig. 4 will be mentioned below.) The experimental proof that the 5.964-Mev group belongs to $Cm^{242}$ lies in the invariance of its abundance relative to the principal groups of $Cm^{242}$. The data are summarized in Table I where the constancy of this ratio is apparent,

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[5] Hollander, Perlman, and Seaborg, "Table of Isotopes," Revs. Modern Phys. **25**, 469 (1953). The values as shown in the "Table of Isotopes" are used here, although some have since been revised.

[6] Thompson, Street, Ghiorso, and Reynolds, Phys. Rev. **80**, 1108 (1950).

[7] Huizenga, Manning, and Seaborg, *The Actinide Elements* [McGraw-Hill Book Company, Inc., New York (to be published)], National Nuclear Energy Series, Plutonium Project Record, Vol. 14A, Chap. 20.

whereas the ratios of this group to those ascribed to $\mathrm{Cm}^{243}$ and $\mathrm{Cm}^{244}$ undergo considerable variation.

Two other possible groups have shown up (Figs. 4a and 4b) at 6.006 and 6.030 Mev, but these cannot be assigned definitely to either $\mathrm{Cm}^{242}$ or $\mathrm{Cm}^{243}$. Further work will be necessary to establish these groups definitely and to make assignments. These groups will be mentioned further under the discussion of the decay schemes.

### Alpha Spectrum of $\mathrm{Cm}^{243}$

The curium preparations which gave rise to the spectra shown in Fig. 4 were made by neutron irradiation of mixtures of americium isotopes and of curium isotopes which in turn had been prepared from americium. The different isotopic compositions obtainable are a function of the starting materials, the neutron capture and fission cross sections, the intensity and duration of irradiation, and the time following irradiation. These factors as applied to the preparation of curium isotopes will be discussed by others.[8]

The objective of comparing the spectra of the different curium preparations is to assign the several groups by comparing abundances. We have already seen that the alpha group at 5.964 Mev follows the abundances of main $\mathrm{Cm}^{242}$ groups and is assigned accordingly as a component of the $\mathrm{Cm}^{242}$ spectrum. Also, groups at 5.755 and 5.797 Mev (Fig. 3) were attributed to $\mathrm{Cm}^{244}$ because the particular sample was prepared in such a way as to make $\mathrm{Cm}^{244}$ and not $\mathrm{Cm}^{243}$.

It remains now to assign groups at 5.985, 5.777, and 5.732 Mev which appear in Fig. 4.

The pertinent numerical data of Figs. 3 and 4 as well as others not shown in graphical form are summarized in Table II in the form of ratios of abundances for the various groups. The group at 5.777 Mev is the most prominent and is used as the reference. Within

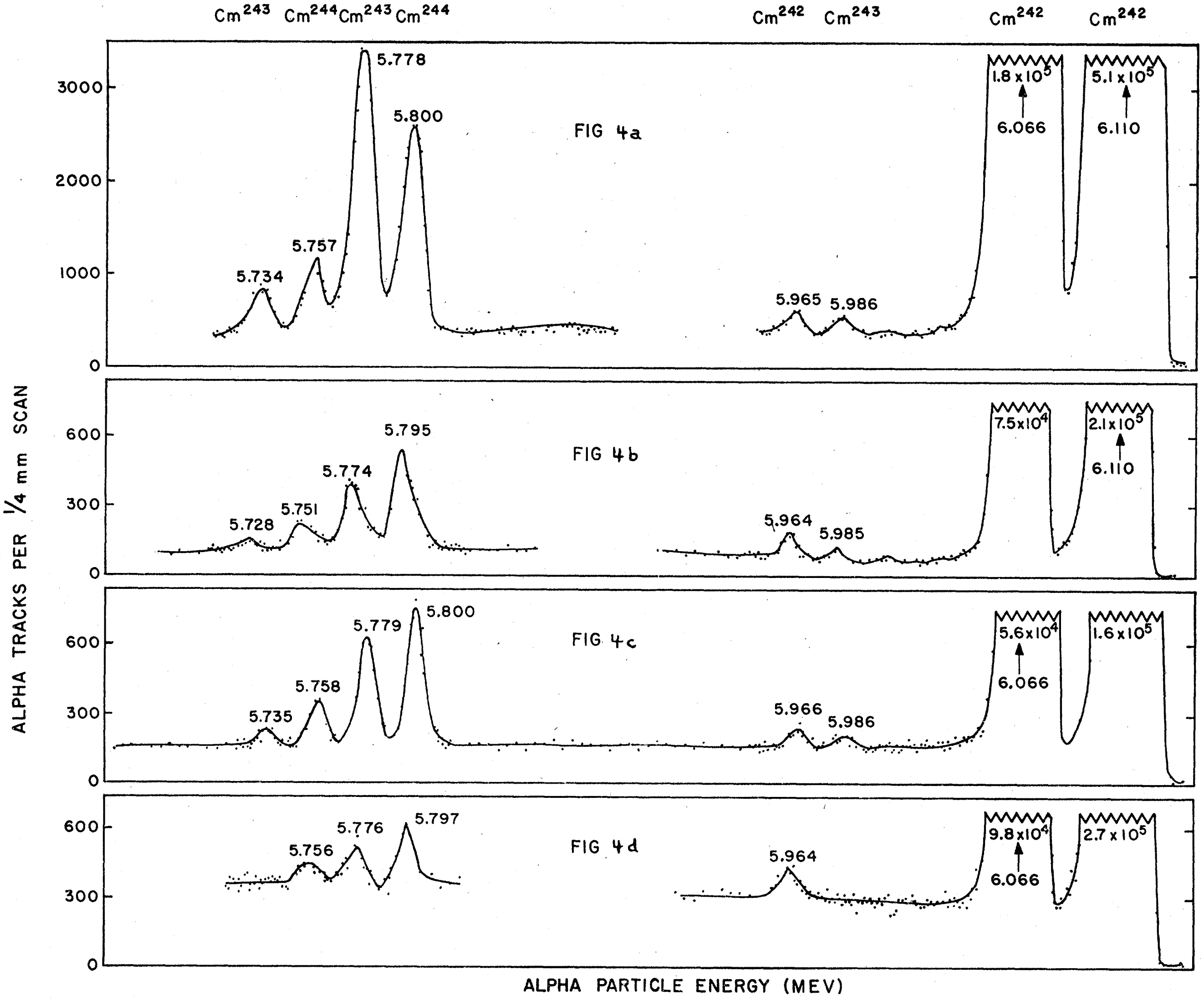

FIG. 4. Alpha-particle spectra of $\mathrm{Cm}^{242}$, $\mathrm{Cm}^{243}$, $\mathrm{Cm}^{244}$ isotopic mixtures.

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[8] Thompson, Barrett, Reynolds, and Higgins (to be published).

TABLE I. Assignment of alpha group at 5.964 Mev to $Cm^{242}$.

| Experiment number / Abundance relative to: | Fig. 4a | Fig. 4b | not shown (No. 105) | Fig. 4c | Fig. 4d |
|---|---|---|---|---|---|
| $Cm^{242}$ groups (at 6.110 and 6.066 Mev) | 0.033% | 0.035% | 0.035% | 0.036% | 0.037% |
| $Cm^{244}$ groups (at 5.798 and 5.755 Mev) | 8% | 16% | 14% | 11% | 53% |
| $Cm^{243}$ group at 5.777 Mev | 8% | 30% | 32% | 18% | 107% |
| Energy of alpha group | 5.965 | 5.964 | 5.963 | 5.966 | 5.964 |

the limits of error in the measurements the ratios of the three groups among themselves are constant.

On the contrary, the ratio in abundance of the 5.777-Mev group to the main group of $Cm^{242}$ varies by almost a factor of 10 and varies by an even larger factor with respect to the main group of $Cm^{244}$. On this basis alone it is probable that all three groups belong to $Cm^{243}$. Other evidence will be discussed presently.

## IV. GAMMA-RAY SPECTRA AND DECAY SCHEMES

### Decay Scheme for $Cm^{242}$

Up to the present study only two alpha groups of $Cm^{242}$ were known, the most abundant leading presumably to the ground state of $Pu^{238}$ and the other to an excited state of about 44 kev, taken to be the first excited state.[2] The $L$- and $M$-shell conversion electrons corresponding to the 44-kev transitions were measured by O'Kelley,[9] and these were seen in the requisite quantity to account for all of the $\alpha_{44}$ transitions[10] of $Cm^{242}$ by Dunlavey and Seaborg[11] using the photographic emulsion technique which indicates alpha particle-conversion electron coincidences. Coincidences between alpha particles and conversion electrons of the proper energy were also measured by electronic means by Prohaska.[12] Finally, conversion electrons which can be assigned to this transition have been observed in high abundances by Freedman, Jaffey, and Wagner[13] in the beta decay of $Np^{238}$.

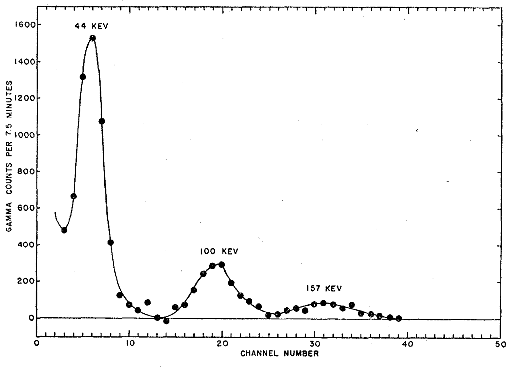

FIG. 5. Gamma-ray spectrum of $Cm^{242}$.

The absence of a gamma ray of comparable abundance to the conversion electrons has been known generally from the low gamma activity of $Cm^{242}$ and this necessarily implies a high conversion coefficient. Goldhaber and Sunyar,[14] in their classification of excited states of nuclei, and Horie and coworkers,[15] independently have stated the generalization that the first excited states of even-even nuclei have spin 2 and even parity. This thesis has been analyzed further by Scharff-Goldhaber[16] who also discussed its implications on the shell model of nuclear structure. The gamma-ray transitions from these states ($2+ \rightarrow 0+$) are of the $E2$ type. Gellman, Griffith, and Stanley[17] have calculated $L$-shell conversion coefficients for $E1$, $M1$, and $E2$ transitions, and from their data we have estimated for the case in question coefficients of 1.5, 60, and 600 for $E1$, $M1$, and $E2$, respectively.

We have measured with a scintillation counter spectrometer the gamma-ray spectrum of $Cm^{242}$ both in samples fairly pure isotopically and in mixtures of curium isotopes. Radiation corresponding to $L$ x-rays was found in abundance, but the actual quantity was not calculated because of the large uncertain attenuation loss in the particular arrangement employed. In much lower intensity were found photons of $44 \pm 3$, $100 \pm 2$, and $157 \pm 2$ kev attributable to $Cm^{242}$. One such spectrum is shown in Fig. 5. The intensity of the 44-kev gamma ray was 1 per $2.4 \times 10^3$ total alpha particles of $Cm^{242}$. Since the first excited state is populated by only 26 percent of the alpha disintegrations, the corresponding yield of gamma rays from this state is 1 per 620 which should represent the *total* conversion coefficient. According to Dunlavey and Seaborg,[11] 83 percent of the electrons seen were $L$-conversion electrons and 17 percent from the $M$, $N \cdots$ shells. Applying this correction, the experimental $L$-shell conversion coefficient becomes 520 which is in good agreement with the theoretical expectations for an $E2$ transition. We can, therefore, have good confidence that the first excited state of $Cm^{242}$ does indeed have spin 2 and even parity. This is indicated in the decay scheme (Fig. 6).

The photon of approximately 100 kev would not be distinguishable from a $K$ x-ray of plutonium, but such an assignment is fairly well ruled out by intensity

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[9] G. D. O'Kelley, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-1243 (unpublished).

[10] The designation $\alpha_{44}$ refers to the alpha particles leading to the 44-kev excited state of $Pu^{238}$; $\alpha_0$ refers accordingly to the alpha transition to the ground state.

[11] D. C. Dunlavey and G. T. Seaborg, Phys. Rev. **87**, 165 (1952).

[12] C. A. Prohaska, Ph.D. thesis, University of California Radiation Laboratory Report UCRL-1395 (unpublished).

[13] Freedman, Jaffey, and Wagner, Phys. Rev. **79**, 410 (1950).

[14] M. Goldhaber and A. W. Sunyar, Phys. Rev. **83**, 906 (1951).

[15] Horie, Umezawa, Yamaguchi, and Yoshida, Progr. Theoret. Phys. (Japan) **6**, 254 (1951).

[16] G. Scharff-Goldhaber, Phys. Rev. **90**, 587 (1953).

[17] Gellman, Griffith, and Stanley, Phys. Rev. **85**, 944 (1952).

TABLE II. Assignment of alpha groups at 5.985, 5.777, and 5.732 Mev to $Cm^{243}$.

| Abundance ratios \ Experiment number | Fig. 3 | Fig. 4a | Fig. 4b | not shown (No. 105) | Fig. 4c | Fig. 4d |
|---|---|---|---|---|---|---|
| Ratio 5.985/5.777/5.732 | | 0.068/1/0.15 | 0.11/1/0.17 | 0.049/1/0.19 | 0.083/1/0.14 | <0.16/1/<0.2 |
| Ratio 5.777/6.110 ($Cm^{242}$) | <0.03 | $5.8\times10^{-3}$ | $1.6\times10^{-3}$ | $1.5\times10^{-3}$ | $2.7\times10^{-3}$ | $4.9\times10^{-4}$ |
| Ratio 5.777/5.798 ($Cm^{244}$) | <0.05 | 1.4 | 0.70 | 0.62 | 0.78 | 0.69 |

considerations. Contrary to the decay scheme of Fig. 6, let us assume that the energy determination of the 157-kev gamma ray is in error and that this gamma ray represents the crossover transition from the state reached by $\alpha_{148}$ to the ground state. On this basis the 100-kev photon could be a $K$ x-ray resulting from the $K$-shell conversion of this gamma ray. The measured abundance of the gamma ray is $2.7\times10^{-5}$ relative to total $Cm^{242}$ alpha particles, and the abundance of $\alpha_{148}$ is $3.5\times10^{-4}$. The $K$-shell conversion coefficient of this gamma ray would then be 12 and according to the calculations of Rose, Goertzel, and Perry[18] this conversion coefficient would correspond to an $E6$ transition.[19] Such a transition in this case can be ruled out on a number of grounds including the lifetime of the state. Furthermore, if the conversion coefficient is 12, the $K$ x-rays should be several times more plentiful than observed. (The 100-kev photon is found in abundance $6\times10^{-5}$.) We therefore consider the ~100-kev photon to be a gamma ray in cascade with the 44-kev gamma ray giving a state 144 kev above the ground state which is in agreement with the existence of the alpha group $\alpha_{148}$. This gamma ray is probably the same as the 103-kev gamma ray reported by Freedman, Jaffey, and Wagner[13] in the decay of $Np^{238}$.

In attempting to assign a multipolarity to the gamma-ray transition from the second excited state to the first and from this to deduce the spin and parity of the second excited state, we have recourse only to an estimation of the conversion coefficient for the ~100-kev gamma ray. Its abundance relative to the 44-kev gamma ray (Fig. 5) is 0.15 and as already indicated there is one 44-kev gamma ray per 2400 alpha particles; therefore, the abundance of the ~100-kev gamma ray is $6\times10^{-5}$ relative to the total $Cm^{242}$ alpha particles. The abundance of $\alpha_{148}$ is $3.5\times10^{-4}$ from which it follows that the conversion coefficient is 5. The theoretical values of Gellman, Griffith, and Stanley[17] for $L$-shell conversion coefficients do not lend themselves for accurate comparison, since their closest gamma-ray energy was ~150 kev and the change of conversion coefficient with energy in this region is rapid. If we use measured conversion coefficients[20] for $E2$ transitions of $Th^{228}$ (83 kev) and $Th^{230}$ (68 kev) to fill out the curve of calculated values[17] the expected value for 100 kev is about 7. The calculated value for an $M1$ transition would be about 5 for this energy while for an $E1$ transition it would be about 0.1. Because of the uncertainties already mentioned in the present comparisons, values within a factor of 2 of each other should be considered in agreement so that we may assign this transition to the categories $E2$ or $M1$.

On this basis alone the second excited state of $Pu^{238}$ would be 4+, 2+, 0+, 3+, 1+. We can rule out the odd states because of the rule mentioned that the odd states of an even-even alpha emitter must have odd parity. Of the other three possibilities we favor 4+ from the synthesis of a number of arguments based on: (1) the absence of a crossover transition to the ground state; (2) a detailed examination of the beta and gamma spectrum of $Np^{238}$; (3) analogies with the excited states of other even-even nuclei for which interpretations are more definite; and (4) the agreement of the energy of this state with the expectations for a 4+

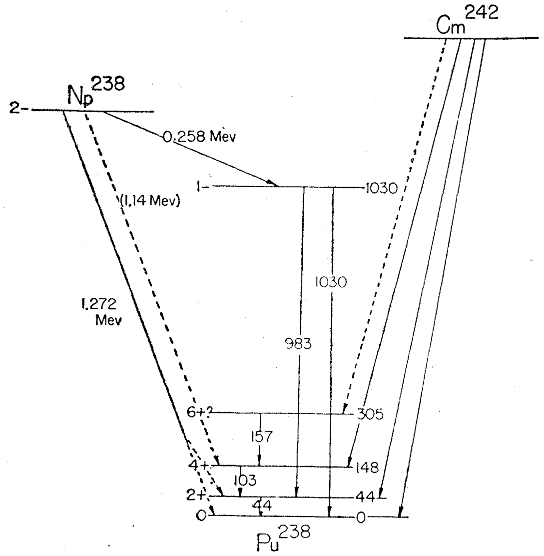

FIG. 6. Decay scheme for $Cm^{242}$ and $Np^{238}$.

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[18] Rose, Goertzel, and Perry, Oak Ridge National Laboratory Report ORNL-1023 (unpublished).

[19] Only electric transitions to the ground state are possible from states reached by an even-even alpha emitter. This follows because all even angular momentum quantum states must have even parity and all odd states have odd parity, and the transitions from such states to the 0+ ground state will involve change in parity from odd states and no change in parity from even states.

[20] For summary of data see reference 5.

state considered as the second rotational state[21,22] of the nucleus. These considerations will be applied generally to the excited states of even-even nuclei at a later date.

As shown in Fig. 5 there is also a gamma ray of 157 kev in low intensity. Its abundance is about one-third that of the $\sim$100-kev gamma ray and consequently is found in abundance $2.7\times 10^{-5}$ of the total alpha disintegrations. As placed in Fig. 6, this gamma ray represents a transition from a state 305 kev above ground state to the excited state at 148 kev, and it is listed as an $E2$ transition. The reasons for this assignment will appear shortly. For an $E2$ transition of 157 kev, the total conversion coefficient should be about 2, and, therefore, the state from which it arises should be populated to the extent of about $8\times 10^{-5}$. Such an alpha group (5.810 Mev) is just on the verge of detection but could not have been seen in the present experiments in any case because the best samples contained Cm$^{244}$ and the principal groups at 5.798 Mev would have obscured it.

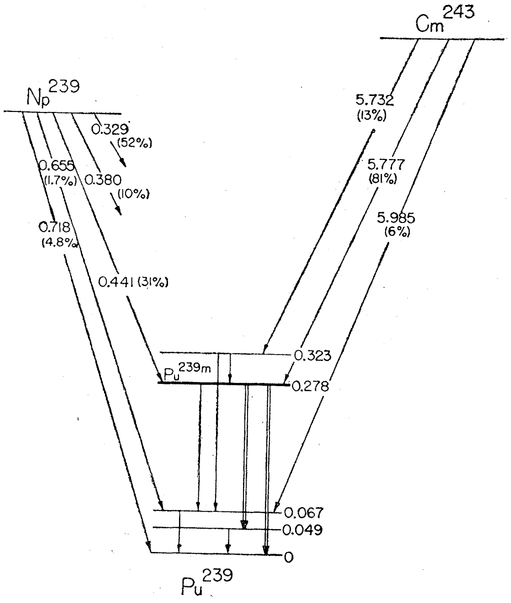

FIG. 7. Decay scheme for Cm$^{243}$ and Np$^{239}$.

Without conclusive justification at present, we shall hypothesize that the most prominent states reached by an even-even alpha emitter among the heaviest elements are states of even angular momentum and that these states represent some configuration of the nucleus as a whole, let us say rotational states. As pointed out by Bohr[21] and by Rasmussen,[22] such states for a nonspherical nucleus should lie at energies proportional to $l(l+1)$. The ratio of energies of the second rotational state to the first would therefore be 3.3 and the third to the first would be 7.0. Applying these factors to the value 44 kev, we arrive at 147 and 308 kev for the second and third rotational states in excellent agreement with energy levels already deduced from alpha- and gamma-ray spectra. In another publication[23] it will be shown that similar agreement is found for other nuclides in this region which have different energy level spacing.[24] However, despite this agreement with theory for the relative energy spacing of the "rotational" levels, the absolute values are not in agreement with reasonable moments of inertia considering the nucleus as a rigid rotator.

If we consider the first three excited states of Fig. 6 to be special in the sense that they represent some quantized behavior of the nucleus as a whole, the question arises as to the existence of other low-lying levels representative of some other mode of excitation. The only indication for such states is the tenuous evidence for two very rare alpha groups at 6.006 Mev and 6.030 Mev (see Figs. 4a and 4b). These would lie 106 kev and 81 kev above the ground state. More refined experiments will have to be performed before the existence of these groups in the decay of Cm$^{242}$ can be considered seriously.

The partial decay scheme for Np$^{238}$ and Cm$^{242}$ shown in Fig. 6 makes use of data obtained by Freedman, Jaffey, and Wagner[13] on Np$^{238}$. The interpretation of these data is conditioned by the energy levels arrived at from the Cm$^{242}$ alpha decay. The principal virtue of the scheme shown is that it is the simplest one which conforms with information now available.

It will be noted that a new $\beta^-$ group is postulated leading to the 148-kev level. This group is needed because of the measurement[13] of coincidences between "hard beta radiation" and an $\sim$100-kev photon. The beta group with end point at 1.272 Mev is taken to go either to the 44-kev state or the ground state, or it may consist of two groups going to both of these levels.

The spin and parity assignments were arrived at from the following arguments: From the alpha-decay data for Cm$^{242}$ already discussed it is almost sure that there are low-lying states of Pu$^{238}$ with designation $0+$, $2+$, and $4+$. The high-energy $\beta$ group (or groups) of Np$^{238}$ has an $ft$ value of 8.5,[13] therefore the ground state of Np$^{238}$ most likely has odd parity as would be expected from the shell model. If this is established, then the high-lying excited state of Pu$^{238}$ must also have odd parity since the $ft$ value for the low-energy beta group could fall into the "allowed" category. The spin values were selected to conform with the selection rules and to account for the near equality in abundance of the two high-energy gamma rays. It is of interest to note that other odd-odd beta emitters (e.g., Ac$^{228}$) with sufficient decay energy also have pairs of hard gamma

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[21] A. Bohr and B. R. Mottelson, Phys. Rev. **89**, 316 (1953).

[22] J. O. Rasmussen (private communication).

[23] F. Asaro and I. Perlman (to be published).

[24] Dr. Aage Bohr has kindly sent us the manuscript of a forthcoming publication (A. Bohr and B. R. Mottelson) in which is included a similar analysis of excited states of even-even nuclei.

rays differing by the energy of the first excited state known from alpha decay.

## Decay Scheme for $Cm^{243}$

The alpha-particle spectrum of $Cm^{243}$ thus far determined consists of the following three groups with indicated abundances: 5.985 (6 percent), 5.777 (81 percent), 5.732 (13 percent). If the 5.985-Mev group corresponds to the ground-state transition, the other two groups would lead to excited states of 212 and 258 kev. However, as will be shown, the 5.777-Mev group is in coincidence with a gamma ray of 278 kev so that the highest energy group seen (5.985 Mev) cannot lead to the ground state. In addition to the 278-kev gamma ray, one at 226 kev was found and a photon of 104 kev believed to be a plutonium $K$ x-ray. All of these photons were attributed to $Cm^{243}$ rather than to $Cm^{242}$ from abundance arguments. Because it is highly likely that the spectrum for $Cm^{244}$ is much like that for $Cm^{242}$, we do not believe that these gamma rays originate from the small amount of $Cm^{244}$ present. Other evidence will be given presently.

The highly complex beta spectrum of $Np^{239}$[20] indicates strongly that there are a number of low-lying excited states of $Pu^{239}$. Limitations of sample intensity of our $Cm^{243}$ preparations are such that alpha and gamma transitions even in moderate abundances could have been missed. The best that can be done at present is to attempt to define the decay energy of $Cm^{243}$ and to reconcile the two abundant gamma rays with the alpha spectrum. The most likely arrangement of these data is shown in Fig. 7 along with a partial beta-decay scheme for $Np^{239}$ involving common levels. Gamma rays observed both from the beta decay of $Np^{239}$ and the alpha decay of $Cm^{243}$ are indicated by double lines. Figure 8 shows, on the same plot, a portion of the gamma-ray spectrum measured with a scintillation spectrometer and the gamma-ray spectrum which is in coincidence with the most abundant alpha group of $Cm^{243}$ (5.777 Mev). The latter measurement was made by setting the magnet field to focus the alpha group on a scintillation detector and using this pulse to trigger the gamma-ray spectrometer. It is seen that the 226-kev gamma ray and 278-kev gamma ray are both in coincidence with the alpha group. (The energies of the gamma rays as indicated are the averages from a number of measurements.) The data are in agreement with the work of Graham and Bell[25] who showed that these two gamma rays originate from the $10^{-9}$-sec metastable state of $Pu^{239}$. As shown in Fig. 7, these gamma rays decay by parallel paths, one leading to the ground state and another to an excited state of 49 kev. Graham and Bell also found a 210-kev gamma ray from the 277-kev level which leads to a 67-kev excited state. We could not measure this gamma ray but did find a low-intensity alpha group which undoubtedly leads to the 67-kev state because its energy is almost exactly 210 kev higher than the group leading to the 278-kev state. The gamma rays shown from the 323-kev state could not be observed in the present study because of the relatively low population of this state by alpha decay, but gamma rays corresponding to transitions from this state have been observed by others[25–27] following the beta decay of $Np^{239}$.

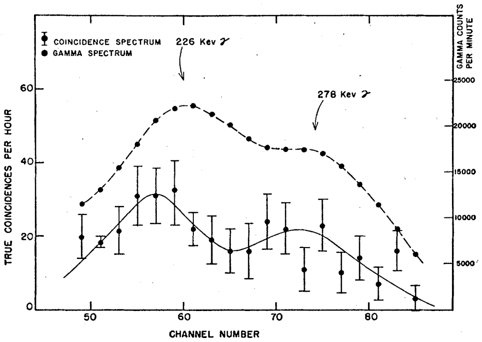

FIG. 8. Alpha-gamma coincidence spectrum of $Cm^{243}$.

It will be noted that the highest energy alpha group measured is in fairly low abundance and that no alpha groups have been observed leading to the two lowest-lying states. These facts are not in accord with unadorned alpha-decay theory which favors highest energy transitions. However, there is ample evidence that the type of spectrum noted for $Cm^{243}$ is the rule rather than the exception for alpha emitters with odd nucleons.

It was mentioned above that the 104-kev photons are probably plutonium $K$ x-rays arising from the internal conversion of the 278-kev and 226-kev gamma rays of $Cm^{243}$. Evidence for this assignment was obtained by setting the gamma-ray scintillation spectrometer for this energy and recording the alpha-gamma coincidences as the magnet current of the spectrograph was varied. Coincidences appeared when the main $Cm^{243}$ group (5.777 Mev) was focused on the detector and not with either of the $Cm^{244}$ groups. The 104-kev photon is therefore in coincidence with the same alpha group which gives rise to the 278-kev and 226-kev gamma rays and is most reasonably handled by assuming it to be a plutonium $K$ x-ray arising from the internal conversion of these gamma rays.

## Decay Scheme for $Cm^{244}$

The only information regarding the decay scheme of $Cm^{244}$ is contained in the observation of two alpha groups differing by 42.5 kev and in abundance ratio of 3 to 1. The prevalence of this structure for even-even alpha emitters in this region makes it highly likely that the decay scheme is similar to that of $Cm^{242}$. On

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[25] R. L. Graham and R. E. Bell, Phys. Rev. 83, 222 (1951).

[26] Freedman, Wagner, Engelkemeir, Huizenga, and Magnusson (private communication).

[27] Tomlinson, Fulbright, and Howland, Phys. Rev. 83, 223 (1951).

this basis the first excited state would be a 2+ state, and the conversion coefficient for the gamma transition would be about 500. Other excited states similar to those of $Cm^{242}$ would be in too low intensity for observation with the preparations now available.

We wish to acknowledge the assistance of Miss Beverly Turner and Mrs. Marjorie Simmons in counting the alpha tracks. We would like to acknowledge the use of the pile facilities and the aid of the personnel of the Atomic Energy Project, National Research Council of Canada, Chalk River, Ontario, Canada, in the irradiations of plutonium, americium, and curium. This work was performed under the auspices of the U. S. Atomic Energy Commission.

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# Slow Neutron Velocity Spectrometer Studies. V. Re, Ta, Ru, Cr, Ga

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New developments in the analysis of data near resonance levels are described. These allow more accurate values of the level constants to be determined from the measured areas above transmission curves and are more general in their applications. The effect of Doppler broadening is also taken into consideration. Transmission data on Re, Ta, Ru, Cr, and Ga are presented. The thermal region of Re is well matched by the relation $\sigma = 5.0 + 15.8E^{-\frac{1}{2}}$. Resonances are found in Re at 2.18, 4.40, 5.92, 7.18, 11.3, 13.1, 17.7, and 21.1 ev. The strong level at 2.18 ev has been studied by the technique of using two samples of different thickness, giving $\sigma_0 = 5700$ barns and $\Gamma = 0.090$ ev. The method of curve fitting was also applied to the transmission dip of the thick sample at 2.18 ev and confirms the values of $\sigma_0$ and $\Gamma$ from the area method. Curve fitting was also used on the 4.40-ev level. New data for Ta in the region above 5 ev are presented. Some transmission dips thought previously to be due to one level have now been resolved into two levels. There are levels at 6.11, 10.2, 13.7, 20.0, 24.0, 35.1, and 38.2 ev. The thermal region of Ru is well matched by the relation $\sigma = 6.4 + 0.39E^{-\frac{1}{2}}$. Resonance levels are observed at 9.8, 15.2, 24.1, and 40.9 ev. The thermal region of Cr is well matched by the relation $\sigma = 3.8 + 0.7E^{-\frac{1}{2}}$. A resonance level is observed at 3800 ev. The nuclear cross section of Ga is given by $\sigma = 7.3 + 0.35E^{-\frac{1}{2}}$ below 5 ev. In addition interference effects occur in the thermal region. Levels were found at 102 ev and 310 ev, with indications of levels at higher energies.

## INTRODUCTION

THE results of previous investigations using the Columbia University slow neutron velocity spectrometer have been presented in a series of earlier papers.[1–3] The present paper is the fifth of a series[2] which presents the results of survey studies of nuclear resonance levels. The improvements and re-evaluation described in the fourth paper[2] apply here. In addition, some new developments in the analysis of data near resonance levels are presented and applied to the data of this report.

### Analysis of Resonance Level Parameters

Techniques for obtaining information about the parameters $\sigma_0$ and $\Gamma$ of a neutron resonance from the experimental transmission curve have been described in previous reports.[1–3] It has been repeatedly pointed out that in the vicinity of a resonance, it is not usually possible to obtain the cross section $\sigma$ from the experimental transmission $T$ by using the formula $T = e^{-n\sigma}$, where $n$ is the number of atoms per cm$^2$ in the sample. The authors find, however, that there seems to be a strong tendency to give direct significance to the curve of $\sigma$ *vs* $E$ obtained in this manner,[4] in spite of the warning to the contrary given in the introduction to this compilation. It should be stressed that such a curve of $\sigma$ *vs* $E$ has little meaning in the region of a resonance level unless the instrumental resolution width is small compared to the level width. In most cases the maximum $\sigma$ obtained in this manner is small compared to the true $\sigma_0$ and decreases with increasing thickness of sample. In certain favorable cases, where $n\sigma_0$ is not large and the level width is not small compared to the resolution width, trial values of level parameters may be chosen and the calculated expected transmission curve (including the effect of the instrumental resolution width) compared with the experimental transmission curve. In especially suitable cases, this may be done in the final stage of the analysis.

As has been pointed out previously, the determination of the level parameters is frequently possible only by an analysis of the area, defined below, of the trans-

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[1] L. J. Rainwater and W. W. Havens, Jr., Phys. Rev. **70**, 136 (1946); W. W. Havens, Jr., and L. J. Rainwater, Phys. Rev. **70**, 154 (1946).

[2] (I) Rainwater, Havens, Wu, and Dunning, Phys. Rev. **71**, 65 (1947); (II) Havens, Wu, Rainwater, and Meaker, Phys. Rev. **71**, 165 (1947); (III) Wu, Rainwater, and Havens, Phys. Rev. **71**, 174 (1947); (IV) W. W. Havens, Jr., and L. J. Rainwater, Phys. Rev. **83**, 1123 (1951).

[3] Rainwater, Havens, Dunning, and Wu, Phys. Rev. **73**, 733 (1948); Havens, Rainwater, Wu, and Dunning, Phys. Rev. **73**, 963 (1948).

[4] *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040 (U. S. Government Printing Office, Washington (1952)).