Radioactivity of Sm^{153} and Eu^{155}†

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The radiations from Sm^{153} and Eu^{155} have been studied by means of a 180-degree, magnetic-focusing, variable-field beta-ray spectrometer-spectrograph using enriched materials. In the disintegration of 47.0-hr Sm^{153}, conversion electrons of three gamma rays of energy, 0.0691, 0.1027, 0.548 Mev, and three beta-ray spectra whose end-point energy and relative intensity were 0.255 Mev, 9 percent; 0.685 Mev, 70 percent; 0.795 Mev, 21 percent, respectively, were observed. A fourth beta-ray spectrum of end-point energy 0.62 Mev and intensity less than 6 percent is postulated. A decay scheme in good agreement with these data and with nuclear shell theory is proposed in which spin and parity have been assigned to all levels. The discrepancy between the magnetic moment measurement and the requirements of shell theory as to the parity of the ground state of Eu^{153} has been resolved in favor of shell theory. In the disintegration of Eu^{155} two beta-ray spectra of end-point energy 0.152 and 0.252 Mev and relative intensity 84 percent and 16 percent, respectively, and conversion electrons of four gamma rays of energy 0.0593, 0.0858, 0.1045, 0.1309 Mev were observed, in agreement with earlier work. Weak conversion-electron lines attributed to gamma rays of energy 0.0187 and 0.1368 Mev were also observed.

I. INTRODUCTION

THE disintegration of Sm^{153} to Eu^{153} has been studied in detail in order to determine whether the case of Eu^{153} represents a violation of the shell-parity correlation[1] as indicated by magnetic moment measurements. Although a number of investigators[2–5] have studied radiations from this 47-hr activity, there has been insufficient agreement to warrant an attempt to assign spin and parity to the observed levels. It appears to be well established that the maximum beta-ray energy is approximately 0.80 Mev, but there has been some doubt as to the number of partial beta-ray spectra associated with the disintegration. The present investigation has shown that the beta-ray spectrum is complex, probably consisting of four components, one of which proceeds directly to the ground state of Eu^{153}.

Previous investigations[2,5,6] of Eu^{155} have indicated the existence of two beta-ray spectra of end-point energies of approximately 0.150 and 0.250 Mev and several gamma rays. In the present investigation all previously reported radiation was confirmed, and in addition two previously unreported gamma rays were also observed. Insufficient data is available for judging between already proposed decay schemes or possible alternate behavior.

II. EXPERIMENTAL METHOD

Measurements were made with the variable-field magnetic focusing beta-ray spectrometer-spectrograph which has been described elsewhere.[7] The instrument was used either as a fixed radius (15 cm) spectrometer or as a fixed field spectrograph. In the latter case the field generated by the current from a storage-battery bank was monitored manually at intervals of six to twelve hours for the duration of the exposure. The sources were investigated both spectrometrically and spectrographically in the same magnet; this procedure resulted in two independent energy determinations of all conversion-electron groups. The discrepancy between these two modes of measurement, as well as that associated with the determination of gamma-ray energies from K, L, and M conversion electrons, has been incorporated in the uncertainty in energy assigned to each gamma ray. Calibration of the instrument was made in terms of the 0.661-Mev gamma ray of Cs^{137}.[8] Measurement of the magnetic field was made through comparison of the alternating voltages generated by two identical coils spinning at opposite ends of the same shaft, one in the field of a permanent reference magnet and the other in the field of the electromagnet. Spectrometer measurements were made with end-window counters filled with alcohol vapor at pressures ranging from 1.2 to 3.5 cm Hg. Windows were made from Formvar films ranging in thickness from 2 to 6 $\mu g/cm^2$, supported by a copper Lectromesh grid. All data were appropriately corrected for window absorption and for electron transmission of the copper grid.

Sources were prepared from enriched, stable samarium isotopes by pile bombardment at the Argonne National Laboratories. The enrichments of the two isotopes used in the investigation were 89.9 percent and 92.1 percent for Sm^{152} and Sm^{154}, respectively. The samarium oxide was dissolved in dilute HNO_3 and was spread uniformly from solution onto 8 $\mu g/cm^2$ collodion backing with the aid of insulin.[9] Sources were 4 cm high and 1 or 2 mm

† Portion of a dissertation submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics at Kansas State College.

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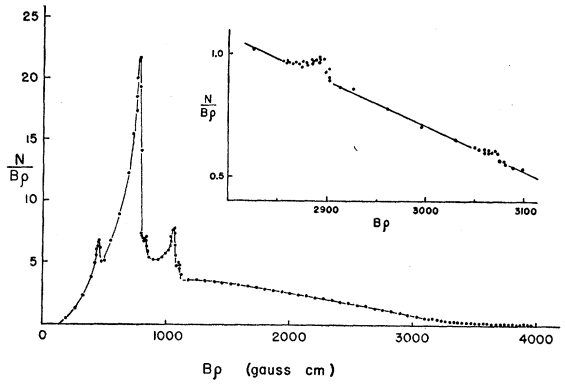

FIG. 1. Electron spectrum of Sm^{153}. From left to right the peaks are due to conversion electrons identified as 69.1-K, 102.7-K, 69.1-L, 102.7-L, -M. K and L conversion electrons of the 548 kev transition are shown in the inset on an enlarged scale.

wide. Source densities ranged from 0.075 to 0.10 mg/cm^2 which resulted in some distortion of low-energy spectra.

All data were taken with statistical accuracies of 1 to 3 percent. The higher statistical error was necessitated by the low specific activity of the Eu^{155} sample.

III. RESULTS FOR Sm^{153}

Figure 1 is a normalized momentum distribution plot of the Sm^{153} activity as observed in the 15 cm radius, 180-deg spectrometer using a 0.2×4.0 cm source. This spectrum has been corrected for the 47.0±0.3 hr half-life determined by monitoring the spectrum for approximately one week. The inset shows an enlargement of the region including the conversion-electron spectrum resulting from the 0.548-Mev transition. Conversion-electron energies were determined photographically as well as spectrometrically. Energy values as determined from both sets of data are summarized in Table I. The K/L ratios were determined from the unnormalized peaks as well as from the normalized areas.[10] It is the average value of the two methods which appears in Table I. This procedure was necessary in view of the self-absorption and backscattering which occurred at low energies in the relatively thick source. The values listed for α_K depend on the proposed decay scheme.

The Fermi plot of the Sm^{153} beta spectrum, which is shown in Fig. 2, shows two distinct changes in slope. Analysis of the plot, assuming an allowed shape for each partial beta-ray spectrum, yields spectra with maximum energies of 0.255±0.010, 0.685±0.005, and 0.795±0.005 Mev, having relative intensities of 9, 70, and 21 percent, respectively. The corresponding $\log_{10}(ft)$ values are 6.0, 6.5, and 7.3. The Fermi analysis was made with the help of tables recently made available by the National Bureau of Standards.[11]

IV. DISCUSSION OF Sm^{153}

The preceding results lead to the main features of the disintegration scheme for Sm^{153} shown in Fig. 3.

The value 3.8 for the K/L ratio of the 69-kev gamma ray is in good agreement with that of 3.5 reported by Mihelich,[12] but is considerably larger than the value of 0.29 reported by Rutledge, Cork, and Burson.[5] Supplementing these data, the K-conversion coefficient of the 69-kev gamma ray has been determined by Sunyar and by McGowan[13] using scintillation spectrometer methods. They found $\alpha_K=3.1$, which tends to confirm the assignment of a mixed $M1+E2$ transition to this gamma ray. The value 6.2 for the K/L-ratio of the 103-kev gamma ray is also in good agreement with the majority of previously reported values. Siegbahn[3] has reported the K-conversion coefficient of the 103-kev gamma ray to be 0.65 as compared with the value of 0.62 determined in this research. McGowan has given $K/(L+M)=3.5$ and $\alpha_K=1.2$ for this gamma ray. These values are in disagreement with the results of the present investigation by a factor of about 2. Nevertheless they remain within the range appropriate to a mixed $M1+E2$ as assigned here. Coincidences between the 69- and 103-kev gamma rays have also been observed.[13] To our knowledge no conversion-coefficient data have been previously reported for the 548-kev gamma ray. The conversion-electron spectrum of this transition is quite weak, a condition which is reflected in the relatively large uncertainties in both α_K and the K/L ratio for this gamma ray. Siegbahn has reported a gamma ray of energy 531 kev as determined by scintillation methods.

A gamma ray of this energy has been reported by both Sunyar and McGowan using similar techniques. Sunyar has also reported a weak gamma ray of energy about 600 kev which has not been observed in the present investigation. Multipole-order assignments were made by comparing experimental values of α_K with those

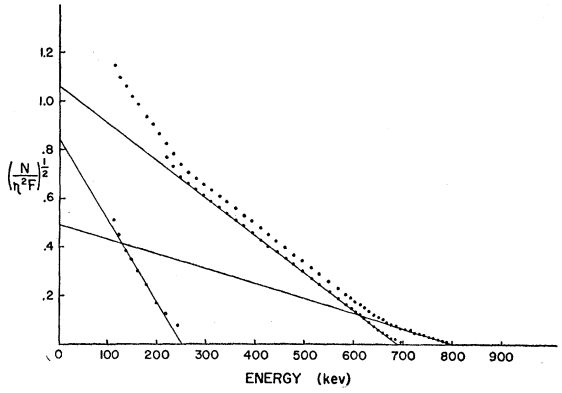

FIG. 2. Fermi plot of the beta-ray spectrum of Sm^{153}.

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TABLE I. Gamma-ray data for Sm^{153}.

Gamma-ray energy (Mev)	Observation[a]	α_K Experimental	α_K Theoretical[b]	K/L Experimental	K/L Empirical[c]	Type
0.0691±0.0002	s, t-6 (K, L)	···	$M1$ 6.0 $E2$ 2.0	$3.8^{+2.5}_{-1.5}$	$M1$ 7.5 $E2$ 0.3	$M1+E2$
0.1027±0.0003	s, t-7 (K, L, M)	0.62 ±0.15	$M1$ 1.3 $E2$ 0.6	6.2±1.5	$M1$ 7.6 $E2$ 0.55	$M1+E2$
0.548 ±0.003	s	0.008±0.004	$M1$ 0.013 $E2$ 0.008	$6.0^{+4.0}_{-2.0}$	$M1$ 7.0 $E2$ 8.0	$M1+E2$

[a] s-observed in the spectrometer; t-6 (K, L)—K and L conversion electrons were observed on 6 different spectrographic films.
[b] See references 14 and 15.
[c] See reference 16.

calculated theoretically by Rose *et al.*[14] and Reitz[15] and by comparing the experimental K/L ratios with the empirical values tabulated by Goldhaber and Sunyar.[16]

Although the spin of Eu^{153} in the ground state is known[17] to be 5/2, no clear-cut parity determination has been made. Magnetic moment considerations place this state as $f_{5/2}$ since the value of μ falls near the "antiparallel" region as computed from the Schmidt model. However, shell-parity correlation prefers the value $d_{5/2}$. The parity of the ground state of Sm_{91}^{153} seems to be clearly odd, there being no evident discrepancy between parity assignments from shell structure theory and experiment in the vicinity of 91 neutrons. The value of $\log_{10}(ft)=7.3$ found for the 0.795-Mev beta ray from the ground state of Sm^{153} to the ground state of Eu^{153} indicates that the transition is first forbidden and requires a parity change. Thus the parity of the ground state of Eu^{153} is even, calling for an assignment of $d_{5/2}$ rather than $f_{5/2}$. Thus the proposed decay scheme assumes the value predicted by shell theory, which results in an internally consistent diagram. The $f_{7/2}$ state for Sm^{153} (91 neutrons) is also in agreement with the shell model. Selection rules for beta decay predict the three experimentally observed spectra to be of the first-forbidden type. The ft values found for these beta spectra are consistent with this assignment. These rules indicate further that the postulated beta-spectrum (dashed line) be higher forbidden than second order in agreement with the experimental difficulty in observing it. Furthermore, calculations from electron intensities and empirical conversion coefficients[13] place the relative intensity of this transition at not greater than 6 percent. The experimental absence of other transitions which might be inferred from the decay scheme is consistent with the spins and parities assigned. The indicated level assignments are precisely those predicted by shell theory, with the first twelve protons in the shell beginning with $A=50$ filling the twelve available $h_{11/2}$ levels.[18] The appearance of the $g_{7/2}$ level instead of an $h_{11/2}$ level is explained by the high pairing energy associated with large j values.

In confirmation, Bannerman,[4] using the integrated spectrum technique,[19] has reported the total energy of the transition from Sm^{153} to Eu^{153} as 0.800±0.020 Mev (plus mc^2).

The K capture of Gd^{153} and the life time of the $S_{1/2}$ state have been added to the scheme for completeness.[20]

Goldhaber and Hill have remarked upon the limited number of long-lived odd-proton isomers between 50 and 82 protons as compared with the relatively abundant odd-neutron isomers in the region of 50 to 82 neutrons. The absence of an excited $h_{11/2}$ level in Eu^{153} may imply that the pairing energy for protons is higher than the pairing energy for neutrons of large j which would account for the absence of odd proton isomers.

V. RESULTS FOR Eu^{155}

The daughter product of 23.5-min Sm^{155} is known[21] to be a long-lived radioactive nucleus Eu^{155} which disintegrates to Gd^{155}. Samarium, having mass number 154 enriched to 92.1 percent, was given a 6-month pile bombardment at the Argonne National Laboratory. The sample was allowed to stand for a period of several

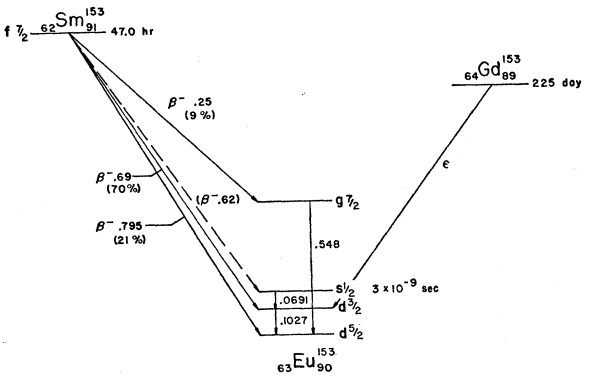

FIG. 3. Proposed disintegration scheme of Sm^{153}. Energies are given in Mev.

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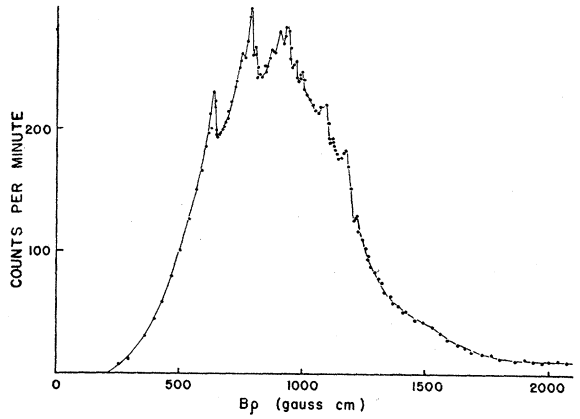

FIG. 4. Electron spectrum of Eu^{155} admixed with some $Eu^{152,154}$ impurity. Thirteen conversion-electron peaks are identified from left to right as 85.8-K, 59.3-L, 104.5-K, 59.3-M, 122.4-K*, 121.2-K*, 85.8-L, 130.9-K, 85.8-M, 104.5-L, 104.5-M, 121.2-L_{Sm}* +122.4-L*, and 121.2-M_{Sm}*+122.4-M*. The starred items are a result of $Eu^{152,154}$. Gamma rays are converted in Gd unless otherwise noted. The unnormalized spectrum is shown.

weeks in order that radiations associated with the disintegrations of the 23.5-min Sm^{155} or the 47-hr Sm^{153} would not be present in objectionable amounts. Figure 4 is the unnormalized spectrum and shows several peaks arising from the decay of $Eu^{152,154}$ even though the mass-spectrographic and chemical analyses showed these materials to be present in very small amounts (0.31 percent). Because of the low activity of the sample, conversion-electron energies were determined photographically. K/L ratios for some of the transitions have been determined as outlined in Sec. III. These values, along with the gamma-ray energies, are listed in Table II.

Figure 5 shows the Fermi analysis of the spectrum indicating three partial beta-ray spectra. Two of these, having maximum energies of 0.152±0.005 and 0.252 ±0.005 Mev and relative abundances of 84 and 16 percent, respectively, are attributed to the decay of Eu^{155}. This is in agreement with other workers.[2] The

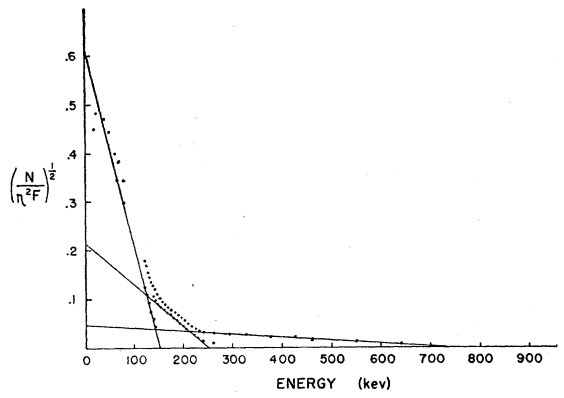

FIG. 5. Fermi plot of the beta-ray spectrum of Eu^{155} and of the Eu^{154} impurity. The two low-energy spectra result from Eu^{155} while the high-energy, low-intensity spectrum results from Eu^{154}.

low-intensity, high-energy spectrum is assigned to the disintegration of Eu^{154}.

VI. DISCUSSION OF Eu^{155}

Four of the gamma rays listed in Table II have been previously reported[2,5,6] in Eu^{155}. Two additional very weak conversion lines which were attributed to gamma rays of energy 18.7 and 136.8 kev by M and K conversion, respectively, were observed in sources enriched in mass number 154 (stable) and in no other, although Sm samples having other isotopic enrichments were studied. Wilson and Lewis have assigned a gamma ray of 15 kev to the disintegration of Eu^{155} on the basis of scintillation measurements. They did not, however, report gamma rays of 59, 131, or 137 kev. Rutledge *et al.* have observed all gamma rays listed in Table II except the 18.7 and 136.8 kev transitions. Both groups named have proposed decay schemes for this activity, but it is felt that there is insufficient evidence reported here to verify either of the schemes.

Assuming a half-life of 1.7 yr, $\log_{10}(ft)$ for the 0.152- and 0.252-Mev beta transities are found to be 6.8 and 8.2, respectively, indicating that they are probably of the first-forbidden type consistent with the requirements of shell-theory.

TABLE II. Gamma-ray data for Eu^{155}.

Gamma-ray energy (Mev)	Observation[a]	K/L
0.0187±0.0001	t-2 (M)	···
0.0593±0.0002	s, t-3 (L, M)	···
0.0858±0.0001	s, t-3 (K, L, M)	$4.0^{+2.0}_{-1.0}$
0.1045±0.0003	s, t-3 (K, L, M)	6.1±1.5
0.1309±0.0004	s, t-3 (K)	···
0.1368±0.0002	t-2 (K)	···

[a] See reference a of Table I.

VII. IMPURITY ACTIVITY

The 0.710±0.020 Mev beta-ray spectrum has been assigned to the disintegration of Eu^{154} on the basis of the relative enrichments of (stable) masses 151 and 153 in the original source material. Mass analysis indicated a ratio of approximately 16/1 in favor of mass number 153 which definitely places this transition with the decay of Eu^{154}. None of the beta spectra associated with the decay of Eu^{152} were observed with this source.

This result completes the assignment of long-lived impurity radiations to the disintegration of Eu^{152} and Eu^{154} on the basis of mass number enrichment in several enriched samarium isotopes.[22]

In summation, gamma rays of energy 121.2±0.3, 243.6±0.7, and 344.2±1.0 kev were assigned to Eu^{152} while a gamma ray of energy 122.4±0.3 kev and a beta-ray spectrum of end-point energy 0.710±0.020 Mev

[22] R. Katz and M. R. Lee, Phys. Rev. **85**, 1038 (1952).

was assigned to Eu^{154}. These energies were determined by spectrometer and by spectrograph. Two further high-energy gamma rays were found in the long-lived europium impurity; one of energy 0.980 Mev by absorption was associated with Eu^{152}, and one of energy 1.17 Mev by absorption was associated with Eu^{154}.

VIII. ACKNOWLEDGMENTS

The cooperation of the Union Carbide and Carbon Corporation, Oak Ridge, Tennessee, in making enriched samarium isotopes available and of the Argonne National Laboratories in bombarding these isotopes is gratefully acknowledged.

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Nuclear Spins and Band Spectra of the Selenium Isotopes

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An investigation of the rotational structure of the $^1\Sigma_u^+ - ^1\Sigma_g^+$ band system of Se_2 in the region 3700–3815A, using separated isotopes of selenium, has made possible the assignment of $I=\frac{1}{2}$ to the nucleus Se^{77}, and the confirmation of $I=0$ for Se^{78} and Se^{80}. At the same time a number of new facts regarding the predissociation and the rotational constants of the Se_2 molecules are established.

I. INTRODUCTION

THE work described here was originally undertaken to resolve a discrepancy[1] that existed between the values of the nuclear spin of $_{34}Se^{77}$ indicated by optical hyperfine structure[2] and by microwave spectra.[3] The former method indicated $I=7/2\pm1$ and the latter $I=\frac{1}{2}$, although with certain reinterpretations[2,4] of the data either result could be reconciled with the other. Under these circumstances it was decided to try the band-spectroscopic method of nuclear-spin determination, since this was the one originally used[5] to establish the value $I=0$ for Se^{80} and is the only one that gives a direct, positive result for a nuclear spin of $I=\frac{1}{2}$ (or $I=0$).

Of the previous analyses of the band spectrum of selenium,[6–10] those of Rosen and of Olsson are the most important. It was clear from the work of Olsson that a separated isotope would be needed to study the rotational structure of the $(Se^{77})_2$ bands, since in the normal isotopic mixture Se^{77} is present to the extent of only 8 percent. [Olsson was able to detect only bands resulting from $(Se^{80})_2$ and $Se^{78}Se^{80}$.] Enriched samples[11] containing as much as 92 percent of this isotope were eventually required to obtain sufficiently accurate intensity measurements on the alternation ratio $(I+1)/I$ in the spectrum of $(Se^{77})_2$. Other samples enriched in Se^{78} and Se^{80} also gave greatly simplified spectra and permitted clarification of certain features of the structure of these bands.[12]

II. EXPERIMENTAL

The sources were electrodeless radio-frequency discharge tubes, this type being chosen principally because of its basic simplicity, freedom from contamination by electrodes, and adaptability to the use of milligram quantities of the elements to be excited. They were straight sealed quartz tubes from 2 to 5 mm in diameter and from 20 to 100 mm long, containing argon gas at several mm pressure and a small sample of metallic selenium, between 2 and 25 milligrams. With this arrangement the entire tube could easily be heated to the desired temperature, if necessary, in the neighborhood of 300°C. To provide better control of the selenium vapor pressure, some of the tubes had side arms which could be heated separately. The external electrodes were made by platinizing the ends of the tubes. Excitation was accomplished by a 60-Mc/sec rf power oscillator, this frequency being chosen because it was high enough to give a satisfactory discharge, but not so high as to require the use of special tubes and circuit components. During operation of the discharge tubes, the power drawn from the oscillator was very small, of the order of 5 or 10 watts.

A large part of the success of this work was due to the availability of enriched isotopes of selenium. In the natural mixture, the principal isotope Se^{80} constitutes

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