Long-Lived Radioactivity of Eu^{152} and Eu^{154}

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The long-lived activity of $Eu^{152,154}$ has been examined by means of a thin-lens coincidence spectrometer and a crystal-summing spectrometer. Decay schemes are given for Eu^{152} and Eu^{154}.

NATURAL europium consists of two isotopes, Eu^{151}(47.77 percent) and Eu^{153}(52.23 percent), which when subject to neutron irradiation transform to Eu^{152} and Eu^{154} by means of the (n,γ) reaction. Both Eu^{152} and Eu^{154} have half-lives of the order of years, with the exact values being uncertain at present.[1–3] In addition, Eu^{152} has a second, 9.2-hour decay, which seems fairly well understood.[4,5]

The decay of the long-lived $Eu^{152,154}$ mixture has been examined by many authors,[5–9] but up to now, no satisfactory decay scheme has existed. Previous workers have identified at least two beta decays and many gamma transitions. These latter are commonly assigned the following energies: 122, 123, 244.3, 336.4, 344, 412, 720, 778, 964, 1086, 1116, and 1402 kev.

In the present work, the beta spectra of the $Eu^{152,154}$ mixture were re-examined, and coincidence studies were made between conversion lines, the conversion lines and the beta spectra, and between the gamma radiations. This information, combined with a knowledge of isotopic assignments drawn from other sources, permits a reasonable ordering of the Eu^{152} and Eu^{154} decay schemes.

APPARATUS

The information gathered here was obtained by means of two separate instruments: a thin-lens coincidence spectrometer and a crystal-summing spectrometer.

The thin-lens coincidence spectrometer consists of two thin-lens spectrometers on a common axis and tube, and with a common source. The individual thin-lens instruments are of conventional design, utilize ring focusing,[10] and are each capable of resolutions of about 2.0 percent. The instrument proper is horizontally mounted in a north-south direction to reduce effects due to the horizontal component of the earth's field. The vertical component is compensated by means of Helmholtz coils. It was found that during operation, the individual focusing coils of the two component spectrometers coupled with each other. The total coupling was less than one percent, and could be easily compensated for in practice. A Garwin circuit[11] of 0.28 microseconds resolving time was used to register coincidences between the Geiger tube detectors of the individual thin-lens instrument.

With a thin-lens coincidence spectrometer of this type, it is possible to utilize either of the component thin-lens instruments in the normal fashion, and to measure coincidences between conversion lines and also between conversion lines and the continuous beta spectrum. A feature of this instrument is the narrowing of coincidence peaks due to source thickness and source size. (A similar effect has been noted for the 180° coincidence spectrometer of Fowler and Shreffler.[12])

Coincidences between gamma rays were observed by means of the crystal summing technique previously described.[13] A thallium-activated sodium iodide crystal, with a 3-mm diameter hole drilled to its center, is coupled to a Dumont 6292 phototube, and the pulses are examined by means of a single-channel discriminator. The gamma-ray spectrum is first examined with the source outside the crystal (i.e., in the normal fashion) and then with the source inserted into the hole. Coincident gamma rays or x-rays, which appear as individual peaks in the first case, add in the second to show a new peak, whose energy is the sum of the contributing peak energies. Similar results are obtained for triple and quadruple coincidences. When this "summing" process takes place, the individual contributing peaks disappear in whole or in part.

The source used in this work was generously provided by Professor J. M. Cork of this university, and is the remains of the material used by Cork, Shull, and Fowler, and is at least five years old. In the thin-lens spectrometer work, the source was evaporated onto a double-layer zapon backing (0.034 mg/cm^2) and covered with a double layer of zapon. In the case of the crystal summing study, the source was enclosed in a Lucite container.

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[3] E. E. Lockett and R. H. Thomas, Nucleonics **11**, No. 3, 15 (1953).
[4] H. B. Keller and J. M. Cork, Phys. Rev. **84**, 1079 (1951).
[5] J. M. Hill and L. R. Shepherd, Proc. Phys. Soc. (London) **A63**, 126 (1950).
[6] Cork, Shreffler, and Fowler, Phys. Rev. **72**, 1209 (1947) **73**, 78 (1948).
[7] F. B. Shull, Phys. Rev. **74**, 917 (1948).
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[10] Keller, Koenigsberg, and Pashin, Rev. Sci. Instr. **21**, 713 (1950).
[11] R. L. Garwin, Rev. Sci. Instr. **21**, 569 (1950).
[12] C. Fowler and R. Shreffler, Rev. Sci. Instr. **21**, 740 (1950).
[13] D. C. Lu and M. L. Wiedenbeck, Phys. Rev. **94**, 501 (1954).

RESULTS

Figure 1 shows the beta spectrum of $Eu^{152,154}$ as measured by the south thin-lens spectrometer. The resolution is 2.2 percent as determined from the K conversion line of the 663-kev transition in Cs^{137}. The conversion peaks are identified. It will be noted that there is a large difference in height between the unresolved 122- and 123-kev peaks and the remainder of the conversion lines. The energies of the unobscured peaks are 244.0 and 343.3 kev in good agreement with previous authors.

Figure 2 is a Kurie plot of the beta spectrum. The high-energy end point is at 1.45 ± 0.05 Mev, followed by a second spectrum with a 0.70 ± 0.03-Mev end point. These values are somewhat lower than those measured by previous workers. (Shull quotes 1.575 ± 0.15 and 0.75 ± 0.015, respectively.) In addition, a third beta spectrum with an end-point energy of 0.330 ± 0.100 Mev is seen. This last beta spectrum must be classified as doubtful, since a second Kurie plot subtraction leads to greatly increased errors. Also, the region in which it is measured contains many conversion electrons, with the accompanying possibility of scattering. In order to minimize this scattering, the beta spectrum has been plotted exclusively from points near the high-energy edge of conversion lines.

Using the thin-lens coincidence spectrometer, coincidences between the conversion lines and the beta spectrum were examined. No coincidences were found between either the 122- or 123-kev L conversion lines and the beta spectrum, or between the 244-kev K conversion line and the beta spectrum. Coincidences were found between the 344-kev K conversion line and the continuous beta spectrum. The coincidence beta spectrum was plotted by fixing the north thin-lens spectrometer on the 344-kev K conversion line and sweeping the south thin-lens spectrometer across the beta spectrum. The resulting Kurie plot is shown in Fig. 3. It will be seen that the 344-kev transition is unambiguously in coincidence with the 0.70-Mev end-point beta spectrum. Shavtvalov[9] has noted similar coincidences, which he attributed to either the 336- or 344-kev gamma-ray line. Examination of our data and Shull's data indicates that the 336-kev gamma ray is not contributing to the coincidences. In fact, we have not seen any such transition.

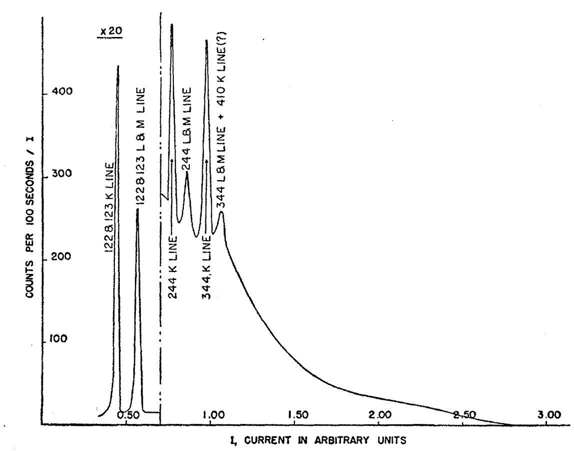

FIG. 1. The beta spectrum of $Eu^{152,154}$. Note the difference in height between the 122+123 conversion peaks and all other peaks.

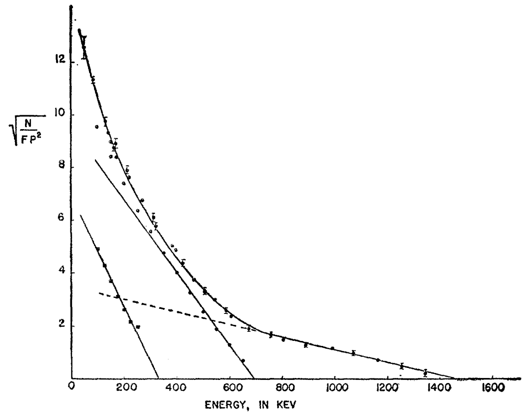

FIG. 2. Kurie plot of the beta spectrum of $Eu^{152,154}$.

Coincidences were then sought between conversion peaks. Either the 122- or 123-kev gamma ray (unresolved in this work) was found to be in coincidence with the 244-kev transition. No other coincidences were noted. This disagrees with certain previous workers, but has been confirmed by the crystal summing data.

The crystal summing data with the principal peaks identified are shown in Fig. 4. Due to the Lucite source container, a certain amount of the continuous beta spectrum is detected in the "summing" spectrum; this accounts for the increased background. From this data, the following results will be noted:

1. Part of the 122- and 123-kev peak (unresolved) is in coincidence with a 39-kev x-ray from K capture, as indicated by the sum peak at 162 kev on the "summing" spectrum.
2. Part of the 122- and 123-kev peak is *not* in coincidence with an x-ray, since it fails to disappear completely on the "summing" spectrum as would otherwise be required. Hence one of the 122- and 123-kev radiations follows K capture; the other does not. This is further indicated by the fact that no coincidence was observed between the 122- and the 123-kev radiations, a fact confirmed by both instruments.
3. The 244-kev transition appears to be in coincidence with an x-ray. (It disappears completely in the "summing" spectrum, and hence must be in coincidence with something of *low* energy.)
4. The 244-kev gamma ray is in coincidence with 122- or 123-kev gamma ray.
5. The 344-kev gamma ray is not in coincidence with anything (i.e., it is unchanged in the two spectra).

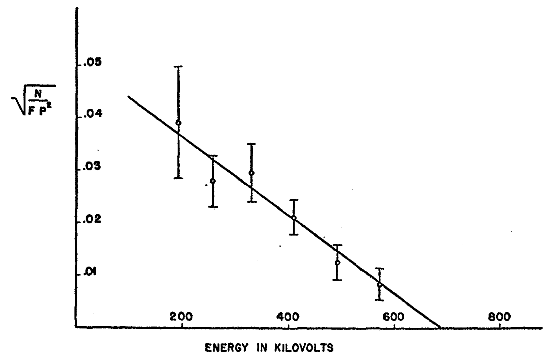

FIG. 3. Kurie plot of the beta spectrum with 0.70-Mev end point plotted from coincidences with the K conversion of the 344-kev transition.

6. An indication of the 410-kev gamma ray is seen, but if any coincidences with it occur, they are obscured by other peaks.

7. A 720-kev peak occurs in the normal spectrum, but any possible coincidences with lower-energy gamma rays are obscured by other peaks in the "summing" spectrum.

8. A large sum peak appears in the "summing" spectrum at $\sim$1140 kev. It will be noted that $720+244+122$ or $123+39\approx 1140$, and that $964+122$ or $123+39$ is also ≈ 1140. In these sums, 39 kev is the K x-ray energy of samarium.

9. A 778-kev line occurs, but it is not in coincidence with anything.

10. A 964-kev line is seen in the normal spectrum. Its partial disappearance in the "summing" spectrum assures that it is in coincidence with something. The broadening at the low-energy base of the 1140-kev sum peak indicates that it is probably in coincidence with an x-ray (also see 8 above).

11. The 1086- and 1116-kev peaks are seen in the normal spectrum, but are unresolved.

12. Sum peaks equal to 1086 kev plus two x-rays and to 1086 plus 122 or 123 kev plus an x-ray are seen.

13. A 1415-kev peak is seen in the normal spectrum.

14. Sum peaks equal to 1415 kev plus an x-ray and to 1415 plus 122 or 123 are seen. No peak equal to 1415 plus 122 or 123 plus an x-ray is seen. Hence 1415 is in coincidence with 122 or 123 or its conversion x-ray, but not with a K-capture x-ray.

ISOTOPIC ASSIGNMENTS

The following isotopic assignments are made by Katz and Lee[14] and shall be adhered to in constructing the decay schemes: the 344, 244, and 122-kev gamma rays belong to the isotope of mass 152; the 1116- and 123-kev gamma rays belong to the isotope of mass 154. This is confirmed by the coincidence measurements made here and by information received privately from Church.[15] Katz's results are based on enriched isotopes, and Church's upon the relative intensity of conversion lines from bombarded and from fission product sources. Church also finds that the 123-kev line is internally converted in gadolinium. In addition, Hill and Shepherd[5] found no positrons from the $Eu^{152,154}$ mixture to one part in four thousand.

This information, combined with the data described above, allows decay schemes to be drawn for Eu^{152} and Eu^{154}.

DECAY SCHEME OF Eu^{152}

The decay scheme for Eu^{152} is shown in Fig. 5. The 122- and 244-kev gamma rays are known to belong to the isotope of mass 152; hence the 244-kev transition must be in coincidence with the 122- and not the 123-kev transition. Since neither the 122- nor the 244-kev gammas are in coincidence with the beta decay, and since they are in coincidence with an x-ray, they are assigned to the K-capture branch of the 152 isotope. The quadruple coincidence of 720, 244, and 122-kev

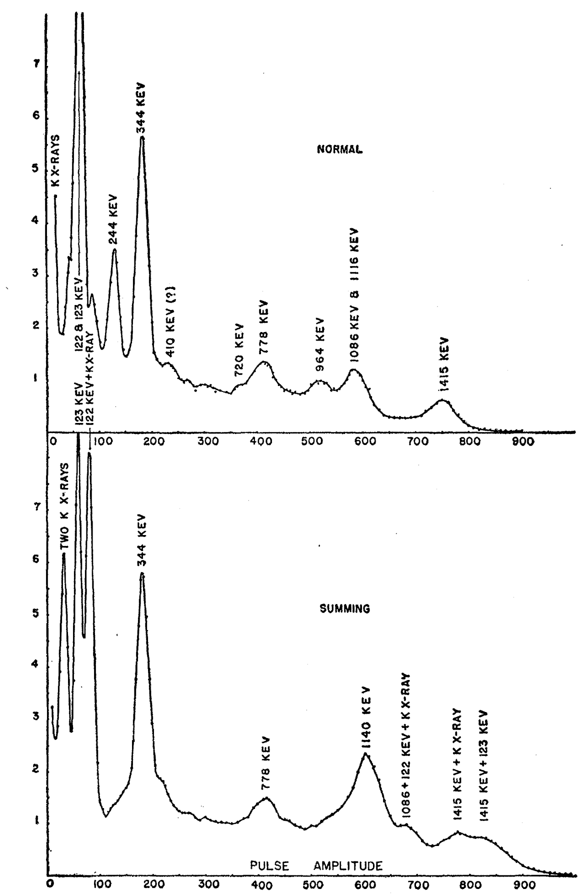

FIG. 4. The normal gamma-spectrum (upper curve) and "summing" spectrum (lower curve) of $Eu^{152,154}$ measured with a crystal summing spectrometer.

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

[15] E. L. Church, Argonne National Laboratories (private communication).

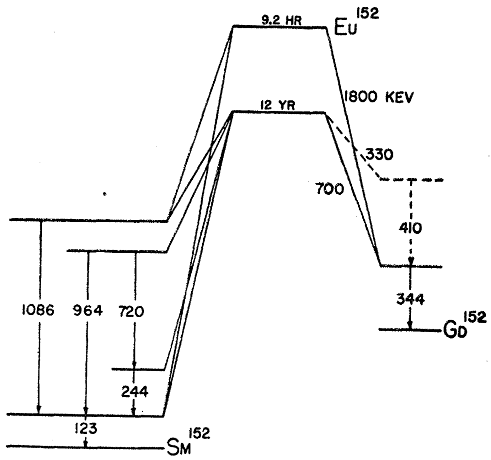

FIG. 5. Proposed decay scheme for Eu^{152}.

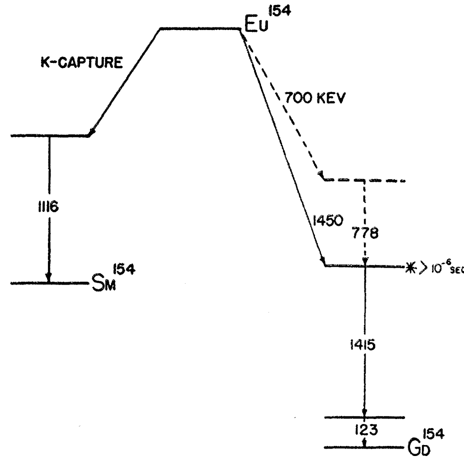

FIG. 6. Proposed decay scheme for Eu^{154}.

gamma rays with the x-ray places them in the position shown. The energy fit of the 964-kev gamma ray, combined with its coincidence with the 122- or 123-kev gamma ray and an x-ray, fits it into the scheme as drawn. Intensity considerations lead to the relative placement of the 122, 244, and 720-kev lines. The 1086-kev gamma ray's position is demonstrated by the triple coincidences of 1086 plus 122 plus an x-ray and 1086 plus two x-rays. (The latter comes about from the high conversion ratio of the 122-kev transition.)

The beta-decay branch is as indicated because the coincidence of the 344-kev gamma ray and the 0.7-Mev end-point beta spectrum has been established, and no other coincidence with the 344-kev gamma ray is seen. The dotted transition between the beta spectrum of end point 300 kev and the 410-kev gamma ray is tentatively proposed because of the energy fit.

The spectrum of the 9.2-hr Eu^{152} has been included in Fig. 5 for the sake of completeness.[16]

[16] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1943).

DECAY SCHEME FOR Eu^{154}

The decay scheme for Eu^{154} is shown in Fig. 6. The coincidence of the 123- and 1415-kev gamma rays has been demonstrated. However no coincidences between the 123-kev gamma ray and either a *K*-capture x-ray or the continuous beta spectrum exists. Since the 123-kev gamma ray is converted in Gd^{154},[15] the beta-decay branch is constructed as shown with the *delay* indicated. The dotted transition is made purely on the basis of energy fit and represents the possibility that the beta spectrum of 0.7-Mev end point actually consists of two spectra of almost identical end points, one occurring in Eu^{152}, the other in Eu^{154}. Both the 778- and 410-kev gamma transitions are dotted in this work. They have been seen only by Cork, Shull, and ourselves using the same source material, and could be due to contamination caused by other rare earths.

Although no coincidences between the 1116-kev gamma ray and an x-ray are seen, they would be obscured by the large 1140-kev "sum" peak. This gamma ray is known to belong to the isotope of mass 154 and does not fit into the beta decay branch. Hence it probably results from *K* capture in Eu^{154}.