Investigation of Conversion Lines in the β -Spectrum of a Eu¹⁵²-Eu¹⁵⁴ Mixture

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Internal conversion lines in the K shells and L and M subshells of Sm^{152} and Gd^{154} were measured with a high resolution β -spectrometer. The ratios of the conversion coefficients were determined for 122 and 123.2 kev transitions. The energy intervals between the conversion lines were measured with high precision.

T HE spectrum of a mixture of the long-lived \mathbf{T}_{Eu}^{152} and Eu^{154} isotopes shows many conversion lines, the most intense of which are due to transitions of Sm¹⁵² and Gd¹⁵⁴ nuclei from the first excited states to the ground states¹. The first excited levels of Sm¹⁵² and Gd¹⁵⁴ are 122 and 123.2 kev respectively². The small energy difference between these levels places the corresponding internal-conversion lines of like shells of Sm and Gd very close together, complicating their separation considerably.

To separate the spectrum of the Sm^{152} and Gd^{154} and to find the ratios of the internal-conversion coefficients in the shells and subshells of each of these elements, it is necessary to employ a β spectrometer of very high resolution. In this investigation we employed for this purpose the prism spectrometer described in Ref. 3, after first increasing its resolution (line half-width 0.04% and in some cases 0.02%)*. In addition, the β -spectrometer was modified to permit accurate measurement of conversion-line energy differences, facilitating considerably the identification of the comversion lines. This β -spectrometer was used to determine the intensity ratios and the energy differences of the strongest conversion lines of Sm^{152} and Gd¹⁵⁴.

A mixture of long-lived Eu¹⁵² and Eu¹⁵⁴ isotopes was obtained by irradiating natural europium with neutrons. After irradiation, the europium was deposited electrolytically on 5-micron thick aluminum foil. A solution of europium chloride in ethyl alcohol was subjected to electrolysis to obtain a series of sources of various thicknesses 10 mm long and 1 mm wide. The source selected from among these was thin enough to provide good resolution and yet give sufficient intensity. This source was approximately several hundredths of a milligram per square centimeter thi

DESCRIPTION OF β -SPECTROMETER

To increase the resolution of the prism β -spectrometer used in this investigation³, it was modified as follows.

1. The spectrometer tubes were shielded. The shield construction was the same as used in the Siegbahn spectro-goniometer⁴. Iron rings and shields (Fig. 1) were placed over the spectrometer tubes to reduce the effects of extraneous magnetic fields on the electrons moving in the tubes. In addition, the shields prevented (to some extent) the iron cover of the spectrometer deflecting magnet from disturbing the axial symmetry of the fields of the lenses.

2. Instead of using a straight slit in the recording equipment, the slit was slightly curved to agree in shape with the image of a line source. To determine the shape of the image, the receiving slit was replaced with a photographic plate, which was exposed to the particles passing through the spectrometer. The source used in this case was a deposit of active RdTh on a 10×1 mm strip of aluminum foil; the spectrometer was tuned to the F line.

3. Some modifications were made to the magnet power supply and to the spectrometer lenses. Power was supplied from a storage battery. The current in the main winding 1 (Fig. 2) and in the lenses 2 was regulated with two slide-wire drumtype resistors (each resistor had a drum approximately 1 m long and approximately 40 cm in diameter). The ratio of the currents in the main winding and in the lenses, and also the current in the auxiliary windings 4 (located above the main winding) and 5 (located on the plates) were varied with ordinary rheostats 6. There was no need to vary this ratio or this current in the auxiliary windings when measuring the group of the peaks corresponding to the conversions in the L and M subshells. Null methods were used to measure the values of all the currents (using a PPTV-1 potentiometer 7 and standard resistors 8).

^{*} The relative half-width of the line is expressed everywhere in this article on a momentum scale.



FIG. 1. General view of prism β -spectrometer: l =plate of magnet yoke; 2 - main winding; 3 - auxiliary winding, placed over the main one; 4 - auxiliary winding on the plate; 5 - demagnetizing winding; 6 screen; 7 - spectrometer chamber; 8 - sylphons (bellows); 9 - diaphragm with round hole; 10 - slit diaphragm restricting the beam vertically; 11 - slit diaphragm restricting the beam horizontally; 12 - tubes; 13 - ground edge; 14 - source; 15 - glass insulator; 16 -source holder; 17 -electrode; 18 -counter chamber; 19 - receiver slit; 20 - counters; 21 - magnetic lens; 22 - iron rings; 23 - lens shields.

The currents in the main winding and in the lenses in small steps, was applied to a suitably-insulated were stabilized by two selenium photocells 9 connected in a bridge circuit and exposed to light reflected from the mirror of sensitive galvanometers 10. A change in current deflects the galvanometer mirror and causes the corresponding photocell, through an amplifier and a relay system 11, to actuate motor 12, which turns the drum of the rheostat so as to return the mirror to its previous position. To reset the equipment to another value of current it is merely necessary to reset the knob of the bridge-circuit potentiometer. The resulting displacement of the mirror causes the rheostat motor to rotate until the required current is established in the circuit.

The electric-bias method was used to measure accurately the energy differences of the conversion lines. A positive voltage, which can be varied

radioactive source (Fig. 1), and all the currents in the β -spectrometer windings were kept absolutely constant. The number of counts n was then plotted as a function of the voltage U applied to the radioactive source. The distance between the conversion peaks could then be read directly in volts from the curve n = f(U). The electric-bias method was used in its time by Lewis and Bowden⁵ to measure the absolute energy difference of monochromatic groups of α -particles. This method was proposed for use in β -spectrometry by Siegbahn^o for accurate determination of the n/e ratio.

Voltage was applied to the radioactive source from dry cells connected in series. The voltage was determined by conventional measurement of the current in a 3.0062 (sometimes 4.0061) megohm wire-wound resistor. This scheme makes it easy to set and measure voltages up to 5 kv with



FIG. 2. Power supply circuit of β -spectrometer.

an accuracy to within 1 volt.

Because it has turned out to be much more convenient to plot small portions of the β -spectrum by clectric bias rather than measure the current in the main windings and in the lenses, this method is now used also to plot spectra of conversion electrons in the determination of the internal-conversion coefficient ratios. It was established first that within the range of bias used in this investigation, the value of the bias does not effect the areas of the conversion lines. To check this, the F line of an active deposit of RdTh was plotted at various values of bias and the line areas, obtained in different measurements, were compared. They proved to be equal, within the limits of statistical dispersion.

L SUBSHELLS OF Sm¹⁵² AND Gd¹⁵⁴

Figure 3 shows a portion of the spectrum of the mixture of Eu¹⁵² and Eu¹⁵⁴, with the lines formed by internal-conversion electrons of 122 and 123.2 kev gamma rays in the L subshells of Sm¹⁵² and Gd¹⁵⁴. The half-width of the lines is approximately 0.05%. The spectrum was plotted under the following conditions: source dimensions 1×10 mm; slit of receiving equipment 1 mm wide, 28 mm long; slit diaphragm 10 (Fig. 1) that limits the vertical divergence of the beam fully open (the only vertical limitation on the beam is diaphragm 9 with a round hole 7 cm in diameter); slit diaphragm 11 which limits the horizontal divergence, cuts a beam 5 cm wide; distance from the source or from the receiving slit to the middle of the corresponding lens 120 cm; aperture ratio of the spectrometer approximately 0.02% of 4π .

The lines could be readily identified by measuring the conversion-line energy differences and comparing these differences with the corresponding energy differences of the electron levels, known from roentgenoscopic measurements⁷.

When measuring the energy difference of two conversion lines (even those that were not adjacent), one line was first plotted, followed directly by the second; the first was then repeated and the two energy differences averaged. To some extent, this eliminated the effect of small systematic changes in the intensity of the deflecting magnetic field, which sometimes occurred and which could introduce an error, albeit small, in the measurement. These changes are apparently due to the slow variation in the spectrometermagnet temperature. The position of the line was established from that of its peak.

Using the measured energy differences of the Sm and Gd conversion electrons and using the electron-level differences given in Ref. 7, it is possible to determine with great accuracy the energy differences of the converting gamma quanta. Thus, for example, the measured energy difference of the conversion electrons from the L_{III} Gd and L_{II} Sm subshells is 1,357 ± 4 ev, from which it follows that the energy difference of the converting gamma quanta in Gd and Sm is 1,292 ev. The error with which the last quantity is determined depends both on the accuracy of our measurements as well as on the accuracy of the data given in Ref. 7.

As can be seen from the drawing, the $L_{III}Sm$ and $L_{II}Gd$ lines did not separate. The distances between these lines and other lines could therefore at first not be established and only the distances from the peak representing the sum of the two lines were measured. To separate the $L_{III}Sm$ and $L_{II}Gd$ lines, the resolution of the β -spectrometer was increased still further in the following manner. An aluminum diaphragm with a slit of 0.5 mm width was placed over the source, thus restricting the width of the source. In addition, diaphragms 10 and 11 (Fig. 1), which limit the divergence of the beam, were so mounted as to form a 4×4 cm



FIG. 3. Portion of β -spectrum of a mixture of Eu¹⁵²

and Eu^{154} isotopes with internal-conversion lines in L subshells of Sm¹⁵² and Gd¹⁵⁴.



FIG. 4. Conversion lines I and I_a of deposits of active RdTh.

square window (the aperture ratio of the instrument was reduced thereby to 0.01% of 4π).

A check on the resolution of the β -spectrometer for the *l* and *la* conversion lines of a deposit of active RdTh (Fig. 4) has shown that under these conditions the instrument line half width was 0.02%.

The L_{III} Sm, L_{II} Gd, and L_{III} Gd lines, plotted with this value of instrument resolution, are shown in Fig. 5. As can be seen, the lines became partly separated. Using graphic line separation, the ratio of the areas (intensities) was found to be L_{III} Sm/ L_{II} Gd = 1.96 ± 0.05. The distance between the lines is 76 ± 2 volts. These data were also used to separate graphically the L_{III} Sm and L_{II} Gd lines on Fig. 3, after which it became possible to obtain the distance between each of the $L_{III}Sm$ and $L_{II}Gd$ lines and the other lines, and also to find the ratios d the internal-convergence coefficients in all the L-subshells.

Table 1 compares the electron-level energy differences E obtained from the roentgenoscopic data given in Ref. 7 with the results of our measurements.

To obtain the ratios of the areas of the Sm and Gd conversion lines, the spectrum of the conversion electrons was plotted both by varying the voltage applied to the source (6 times), as well as by the usual method of varying the current in the principal windings and in the lenses (4 times). The first method of plotting the spectrum yielded the following area ratios: for samarium $L_{II}/L_{I} = 2.25 \pm 0.07$, $L_{III}/L_{I} = 2.26 \pm 0.07$; for gadolinium



FIG. 5. Conversion lines L_{III} Sm, L_{II} Gd and L_{III} Gd, plotted at increased resolution.

TABLE 1

| | Differences in electron energy levels of Sm, ev | | | Differences in electron energy levels of Gd, ev | | |
|---------------|--|---|---|--|---|---|
| | $E_{L_{II}} - E_{L_{I}}$ | $E_{L_{\text{III}}} - E_{L_{\text{III}}}$ | $E_{L_{\text{III}}} - E_{L_{\text{III}}}$ | $E_{L_{II}} - E_{L_{I}}$ | $E_{L_{\text{III}}} - E_{L_{\text{III}}}$ | $E_{L_{\text{III}}} - E_{L_{\text{I}}}$ |
| From Ref. 7 | 426 | 596 | 1022 | 441 | 691 | 1132 |
| From our work | 424 <u>+</u> 2 | 597 <u>±</u> 1 | 1021 <u>+</u> 2 | 443 <u>+</u> 4 | 691 <u>+</u> 4 | 1131±2 |

$$\begin{split} L_{III}/L_I &= 2.80 \pm 0.10, \ L_{III}/L_I &= 2.52 \pm 0.07. \ The \\ \text{second method yielded for samarium } L_{II}/L_I &= 2.20 \\ \pm 0.05, \ L_{III}/L_I &= 2.07 \pm 0.07; \ \text{for gadolinium } L_{II}/L_I \\ &= 2.51 \pm 0.13; \ L_{III}/L_I &= 2.30 \pm 0.13. \ \text{As can be seen,} \\ \text{the ratios of the conversion-line areas obtained by} \\ \text{different methods fluctuate within the limits of} \\ \text{statistical error. The area ratios deduced on the} \\ \text{basis of all the measurements are } L_{II}/L_I &= 2.22 \pm 0.04 \\ L_{III} \ L_I &= 2.16 \pm 0.05 \ \text{for samarium and } L_{II}/L_I \\ &= 2.69 \pm 0.08, \ L_{III}/L_I &= 2.46 \pm 0.06 \ \text{for} \\ \text{gadolinium. It follows therefore that the conversion-coefficient ratios are } L_1:L_{II}:L_{III} &= 1: \\ (2.22 \pm 0.04)_{2}^{\prime} \ (2.16 \pm 0.05) \ \text{for Sm and } L_I:L_{II}:L_{III} \\ &= 1: (2.69 \pm 0.08): \ (2.46 \pm 0.06) \ \text{for Gd.} \end{split}$$

The experimental values of the ratios of the internal conversion coefficients were compared with theoretical values obtained interpolating the data given by Rouse in Ref. 8. The interpolation was performed graphically. To interpolate by energies, the logarithms of the coefficient of conversion ratios were plotted against the logarithm of the transition energy was plotted. To interpolate by atomic number, a plot was made of the logarithm of the conversion-coefficient ratio vs. the atomic number at the transition energy of interest to us. Comparison of the theoretical values of the ratios of the internal-conversion coefficients, obtained in this manner, with the measured values establishes this transition, as expected to be of the electric quadrupole (E2) type for both Sm¹⁵² and Gd¹⁵⁴. For this type of transition, the theoretical values of the conversion-coefficient ratios (with transition energies expressed in m_0C^2 units, k = 0.24) are $L_I:L_{III}:L_{III}$ = 1: 2.3: 2.2 for samarium and $L_I:L_{III}:L_{III}=1$: 2.7: 2.5 for gadolinium. This agrees with the measured ratios of the conversion coefficients.

M-SUBSHELLS OF SAMARIUM

Figure 6 shows a portion of the β -spectrum with the lines formed by the internal-conversion electrons in the *M* subshells of Sm (transition energy 122 kev). By measuring the energy intervals between the *M* lines, and also between *M*



FIG. 6. Internal conversion lines in M subshells of Sm^{152} .

lines and the nearest lines of the L group $(L_{III}Gd)$, it is possible to identify them uniquely, as shown in the diagram. The lines M_{IV} and M_{V} are apparently considerably weaker than those shown on the drawing, and were not noted.

The ratio of the conversion coefficients in the M subshells obtained in this investigation is $M_{II}M_{III}M_{III} = 1$: (3.4 ± 0.1): (3.3 ± 0.2) (average of eight measurements). The striking fact is that conversion occurs principally in the M_{II} and M_{III} subshells. This agrees with the deductions made on the basis of approximate (non-relativistic, without allowances for shielding) calculations of the relative conversion coefficients in the M subshells⁹.

The ratio of the total conversion coefficient in the L subshell to the total conversion coefficient in the M subshell of Sm was measured. The value obtained was $L/M = 4.5 \pm 0.1$ ($L = L_{\rm I}$ $+ L_{\rm II} + L_{\rm III}$; $M = M_{\rm I} + M_{\rm II} + M_{\rm III}$).

MEASUREMENTS OF RATIOS OF CONVERSION COEFFICIENTS IN K AND L SUBSHELLS OF Sm AND Gd.

The K lines of Sm^{152} and Gd^{154} , corresponding to the above transition energies, are shown in Fig. 7. The energy difference of these conversion lines, measured by applying an electric bias to the source, was 2,117 ± 1 ev.

To find the ratios of the coefficient in the Kshells to the total conversion coefficients in the L shells of Sm and Gd, the lines were plotted both by using electric bias, as well as by varying the current in the principal windings. When plotting the groups of K lines or groups of L lines by means of electric bias, the current in the spectrotrometer windings was adjusted to correspond approximately to the energy of the plotted group of lines. The value of the potential difference applied to the source was therefore small in all cases (it should have insured the plotting of a spectrum within the limits of a single group). Using this method of measurement, the correction for the value of the energy interval (expressed in ev) should be introduced when changing from one group of lines to the other.

When plotting the spectrum by varying the current in the main windings, the number of counts of the recording device were plotted vs. the value *l* of this current. Here, therefore, in addition to the usual correction for the width of the pulse interval, a correction was introduced for the non-linear dependence of the spectrometer magnetic field on *l*. The magnitude of this correction could be obtained without difficulty from the known differences between the momenta of the electrons belonging to different lines in the measured groups of conversion lines. In view of the considerable energy difference between the K and L electrons, a correction was also introduced for the difference in their absorption in the counter films. The curves given in Ref. 10 were used to determine this correction. Inasmuch as the film over the windows of the recording installation



FIG. 7. Internal conversion lines in K shells of Sm^{152} and Gd^{154} .

was relatively thin (the counter chamber was separated from the evacuated portion of the spectrometer by a colloidal film 0.23 mg/cm^2 thick and the total thickness of the colloidal film covering the output opening of the first counter and the input opening of the second counter was 0.1 mg/cm^2), this correction amounted to merely 3% of the K/Lratio.

After processing all the measured data, the following values were obtained for the conversion coefficient ratios: for Sm (transition energy 122 kev) $- K/L = 1.76 \pm 0.04$; for gadolinium (transition energy 123.2 kev) $- K/L = 1.51 \pm 0.03$.

In determining the theoretical values of K/L, we employed the K shells conversion coefficients calculated by L. A. Sliv and I. M. Band (private communication), making allowances for the shielding and for the finite dimensions of the atomic nucleus. The L shell conversion coefficients were obtained by interpolation from the data of Ref. 8. The theoretical relationships obtained in this manner for an E2 transition and a transition energy k = 0.24 were K/L = 2.2 for samarium and K/L= 1.9 for gadolinium.

It must be noted that the small number of the computed L subshells conversion coefficients did

not permit sufficiently accurate interpolation. On the other hand the experimentally-obtained K/Lratios are also subject to noticeable systematic errors, owing to the relatively large energy difference between the K and L electrons. These factors apparently explain also the sufficiently large discrepancy between the experimental and theoretical values of the K/L ratios.

1 Slattery, Lu and Wiedenbeck, Phys. Rev. 96, 465 (1954).

2 Cork, Keller, Rutledge and Stoddard, Phys. Rev. 77, 848 (1950).

3 Kel'man, Kaminskii and Romanov, Izv. Akad. Nauk, Ser. Fiz. SSSR, 18, 209 (1954).

4 K. Siegbahn, Ark. for Fys. 4, 223 (1952).

5 W. B. Lewis and B. V. Bowden, Proc. Roy. Soc. (London) **A145**, 235 (1934).

6 K. Siegbahn, Appl. Sci. Res. B4, 25 (1954).

7 Hill, Church and Mihelich, Rev. Sci. Instr. 23, 523 (1952).

8 K. Siegbahn, Beta-and Gamma-Ray Spectroscopy, Amsterdam, 1955, p. 905.

9 E. L. Church and J. E. Monahan, Phys. Rev. 98, 718 (1955).

10 D. Saxon, Phys. Rev. 81, 639 (1951).

Translated by J. G. Adashko

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