PHYSICAL REVIEW VOLUME 91, NUMBER 2 JULY 15, 1953

Gamma-Ray Spectrum of Ionium (Th^{230})†

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(Received March 16, 1953)

The gamma-ray spectrum following the alpha-decay of ionium (Th^{230}) was investigated using a scintillation spectrometer. The following gamma-ray energies (in Kev) and intensities (quanta per disintegration) were observed: 68(0.010); 142(0.0014); 255(0.0005). The 68-kev and 142-kev lines were found to be emitted in cascade; the third apparently represents a transition to the ground state. A level diagram for the product nucleus, Ra^{226}, is suggested.

I. INTRODUCTION

THE gamma-ray spectrum following the alpha-decay of ionium (Th^{230}) has been previously investigated by means of absorption measurements[1-3] and analysis of conversion electrons.[4-7] Further information is supplied by a study of the structure of the alpha-ray spectrum.[8] All observers agree on the presence of a highly converted gamma-ray of approximately 70-kev energy. Probably the most accurate measurement of the energy of this line was performed by Rosenblum and Valadares[5] by analyzing the conversion electrons in a magnetic spectrometer. Three separate conversion lines yielded a gamma-ray energy of 67.8 kev. Higher energy radiation, in the region between 100 and 300 kev, was reported by all the cited investigators; however, no accurate energy measurements could be obtained from the absorption curves, while the internal conversion lines were too weak to be observed. The analysis of the alpha-ray spectrum[7] yielded similar results, showing a line of about 25 percent intensity at 70 kev from the main group leading to the ground state of the product nucleus. There was indication of weaker lines representing transitions to higher excited states; however, neither energies nor intensities could be satisfactorily measured.

It therefore appeared worth while to undertake a new study of the gamma-ray spectrum using the increased sensitivity afforded by the scintillation spectrometer.

II. EXPERIMENTAL PROCEDURE

An Io solution, containing 90 percent Th^{232} and 10 percent ionium was supplied by the Argonne National Laboratory. The gamma-activity of the thorium decay products was negligible in the present experiments (see later discussion). A source yielding 2.8×10^4 disintegrations per second was deposited electrochemically on a thin platinum foil.[9] This thin source was used for calibration of the alpha-emission and absolute determination of the intensities of the lower-energy gamma-ray lines. The weaker line of higher-energy was studied using most of the available material, evaporated on a glass plate. The relative activities of the two sources could be accurately compared by means of the 67.8-kev gamma-ray. The stronger source has an activity of 4.85×10^5 disintegrations per second.

The scintillation spectrometers consisted of the usual NaI(Tl) crystals, 2.1 cm×2.1 cm×1.0 cm in size, carefully selected P-5819 phototubes, linear amplifiers, and single-channel pulse analyzers. For the coincidence experiments, the outputs of the pulse analyzers were fed into a coincidence circuit of 1.8-microseconds resolving time. The delays introduced by the pulse analyzers were found to have no effect on the coincidence efficiency for resolving times of this order of magnitude.

The source was placed at variable distances from the crystals depending on the particular purpose of the investigation. In the coincidence experiments, usually the source was placed between the two crystals, at distances of about 3 mm from either surface. In this arrangement, the solid angle subtended by each crystal was of the order one-fourth of the total; hence high coincidence efficiencies could be obtained. In the experiments designed to determine the absolute intensities of the lines, when sufficient intensity was available, the source was placed at greater distance from the crystal, in order to define better the solid angle subtended by the crystal and minimize the differences in effective solid angles for radiations of different energies, owing to different penetration of the gamma-rays in the sodium iodide. For the determination of the gamma-ray energies, when the knowledge of the absolute intensities was not required, the strong source was placed as close as possible to the crystal, and appropriate absorbers were interposed, to minimize the background due to the high-energy wing of neighboring lines of lower energies.

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† Work supported by the U. S. Atomic Energy Commission.

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[6] C. J. D. Jarvis and M. A. S. Ross, Proc. Cambridge Phil. Soc. **A64**, 535 (1951).

[7] P. Falk-Vairant and J. Teillac, J. phys. et radium **12**, 659 (1951).

[8] Rosenblum, Valadares, and Vial, Compt. rend. **227**, 1088 (1948). Further results quoted by Curie (reference 2).

[9] Battey, Madansky, and Rasetti, Phys. Rev. **89**, 182 (1953). The authors are indebted to Dr. Battey for preparation of this source.

TABLE I. Energies and intensities of the ionium gamma-ray lines.

Energy kev	Pulses in photoelectric peak per sec	Fraction of total solid angle	Efficiency of crystal	Percent of pulses in photoelectric peak corrected for K-radiation escape	Fraction transmitted by absorbers	Quanta emitted per sec	Quanta per alpha
14±1	1310	0.029	1.00	1.00	0.79	57 000	0.117
68±1	351	0.080	1.00	0.91	0.95	5100	0.0105
142±2	38	0.064	0.91	0.98	1.00	667	0.0014
255±3	5.1	0.055	0.51	0.75	1.00	243	0.00050

The resolution of the spectrometer was usually calibrated by means of the 87.5-kev line of Cd^{109}. This line showed a full width of 25 to 28 percent at half-maximum. The relative widths of lines of different energies indicated proportionality to $1/E^{\frac{1}{2}}$ as expected. The energy scale was calibrated using the 46.7-kev line line of RaD, the 87.5-kev line of Cd^{109}, the 238.6-kev line of ThB, the annihilation radiation, and the 661.6-kev line of Cs^{137}. For the last three lines, the recent accurate values of the energies determined by Muller, Hoyt, Klein, and DuMond[10] were used. The scale of the pulse analyzer was linear within the accuracy of the measurements.

The alpha-activity of the electrolytically deposited source was determined by counting the alphas emitted within a small solid angle both with a thin-walled Geiger counter and with a scintillating crystal. Although the thickness of the source was too great for observing a sharply defined range, a sufficient plateau in the integral range curve was found to allow a fairly accurate count of the alpha-emission.

III. ENERGY MEASUREMENTS

The spectral region from 10 to 800 kev was explored for photoelectric peaks due to gamma-ray lines. Three such lines were found, in addition to strong L x-rays due to internal conversion of the 67.8-kev gamma-ray. Of course, the presence of weaker lines cannot be excluded, as, especially in the vicinity of the stronger lines, low peaks would be difficult to observe. Spectra were taken with various filters, to facilitate observation of possible additional lines. An attempt was made to detect the K x-ray of radium at 88 kev, resulting from conversion of the higher-energy lines. For this purpose, the radiation was filtered through 1 g/cm² of Bi in order strongly to reduce the intensity of the 68-kev line from which the 88-kev x-rays could not be well resolved. Indications of the presence of x-rays with an intensity comparable to that of the 142-kev line were found; however, various perturbing effects prevented us from measuring the intensity of this x-radiation with any accuracy.

Before accurate measurements were performed, it was thought that the line near 250 kev might be the 238.6-kev line of ThB due to the thorium impurity in the source. However, careful comparison showed a definite energy difference. Furthermore, it can be calculated that even if the thorium were accompanied by the equilibrium amount of RdTh and its decay products, the intensity of the 238-kev line would be but 1/20 of the observed intensity of the 255-kev line.

The energy of each line was measured under various conditions, by comparison with different calibration lines, and the agreement between the values thus obtained led to the estimate of the possible errors indicated in Table I, where the energies of the three lines are given, together with the intensity data obtained from the experiments described below.

Figures 1 and 2 show the differential pulse-size spectra obtained in two different energy ranges.

IV. INTENSITY MEASUREMENTS

The number of photons emitted in each line was determined by measuring the total number of pulses in the photoelectric peak recorded by the crystal under known geometrical conditions. For the stronger lines at 14 kev (L x-rays), 67.8 kev, and 142 kev, the thin ionium source was placed at 2 or 3 cm from the surface of the crystal. Under such conditions, the solid angle is well defined and not too dependent on the penetration of the radiation in the crystal. For the 255-kev line, the stronger source was used. In order to determine the total number of photons emitted by the source, one must correct for (1) the effective solid angle subtended by the crystal; (2) the efficiency of the crystal, derived from the knowledge of the absorption coefficient in sodium iodide; (3) the percentage of pulses expected to fall within the photoelectric peak, which has been calculated for different energies by Novey;[11] (4) the percentage of iodine K radiation escape, which subtracts pulses from the photoelectric peak; this correction was only applied to the 67.8-kev line, where it is about 10 percent; (5) absorption in 0.188 g/cm² Lucite and 0.015 g/cm² Al interposed between the source and the crystal. The absorption of the L-x-rays was experimentally determined, as it represents an important correction.

Table I summarizes the results, including energies of the lines, measured integral numbers of pulses per second in each photoelectric peak, solid angle for each measurement, and the various correction factors applied. The last column gives the number of photons per disintegration in each of the three gamma-ray

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[10] Muller, Hoyt, Klein, and DuMond, Phys. Rev. 88, 775 (1952).

[11] T. B. Novey, Phys. Rev. 89, 672 (1953).

lines and the number of x-ray photons in the unresolved L group whose average energy is about 14 kev. All values are referred to the stronger source even when actually measured with the weaker source, in order to facilitate comparison.

It is difficult to estimate the errors for these intensity values, as several of the correction factors are inaccurate, being evaluated for normal incidence of the gamma-radiation on the crystal, a condition not always well fulfilled under the large solid angles which had to be used. The absolute intensities of the x-rays, the 67.8 and 142-kev lines should be accurate to about ten percent, the intensity of the 255-kev line within 20 or 30 percent.

V. COINCIDENCE EXPERIMENTS

Gamma-gamma coincidences were investigated using two crystal spectrometers as described before. Either the stronger or the weaker source was used, depending on the intensities of the lines. The solid angle subtended by each crystal was always of the order of one-fourth of the total. Under such conditions, some difficulty was caused by the occurrence of spurious coincidences, owing to one of the higher-energy gamma-rays undergoing Compton scattering in one crystal and the scattered photon being detected by the other crystal. To distinguish such spurious coincidences from those due to emission of two gamma-rays in cascade, one of the spectrometers was set to cover the pulse-size range corresponding to one of the ionium lines, while a differential spectrum of the coincident pulses in the other channel was determined. If this spectrum showed a peak at the proper position for another of the ionium lines, the latter was known to be in cascade with the first. All such combinations of the three gamma-ray lines and of the L x-ray were tried. The only definitely established coincidences are those between the 142-kev line and either the 67.8-kev line, or the x-rays representing its internal conversion. The latter are much easier to observe on account of the higher intensity. Absence of coincidences for the combinations (255–142) and (255–67.8) seems decisive, since these should have been readily observed if the corresponding gamma-rays were in cascade.

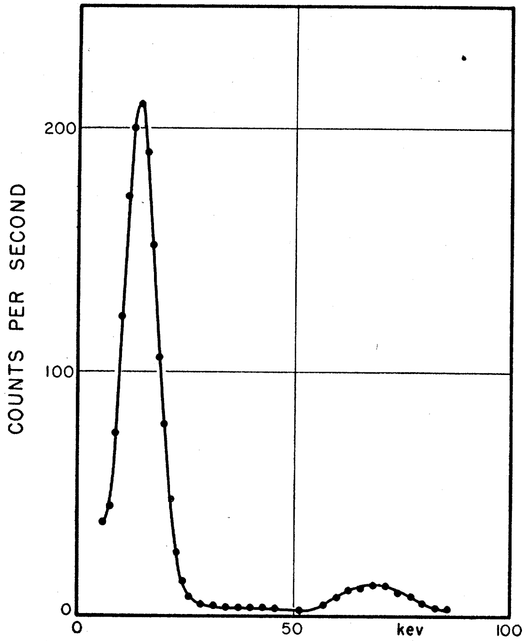

FIG. 1. Differential pulse-size spectrum in the region 0–80 kev (without absorber) showing x-rays at 14 kev and gamma-ray at 68 kev.

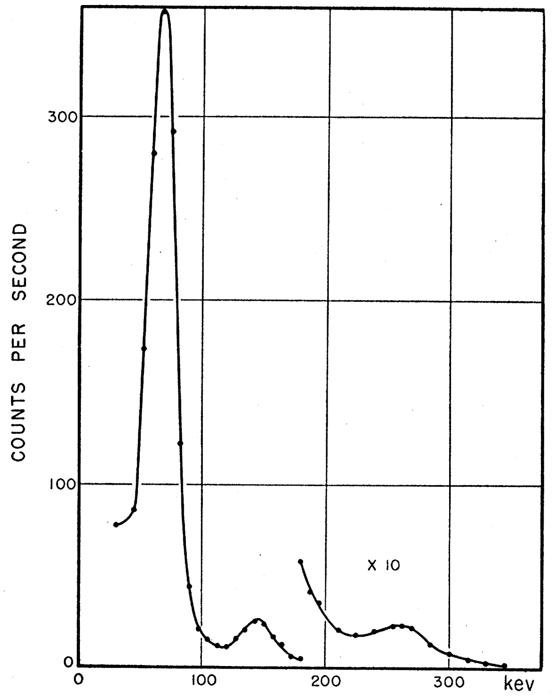

FIG. 2. Differential pulse-size spectrum in the region 0–300 kev (without absorber) showing gamma-rays at 68, 142, and 255 kev.

The number of coincidences between the 142-kev gamma-ray and the x-rays representing the internal conversion of the 67.8-kev line was of the order of, but somewhat smaller (perhaps one-half) than expected from an estimate of the solid angles involved, efficiencies of the crystals, conversion coefficients of the 67.8-kev line in the three L sublevels, and their respective fluroescent yields. Since the latter quantities are not well known, the discrepancy cannot be considered significant. It was thought that lack of the full number of coincidences expected might be due to a half-life of the intermediate level of the same order of magnitude as the resolving time of the coincidence circuits. To test this assumption, the coincidences were analyzed by means of an oscilloscope. The sweep was triggered by the signals representing the 142-kev line, while the pulses from the channel recording either the 67.8-kev

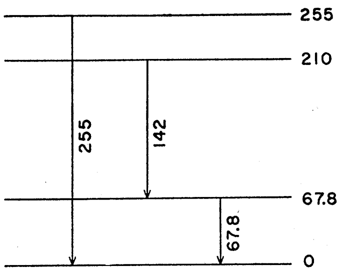

FIG. 3. Suggested level diagram for the Ra^{226} nucleus.

line or the x-rays were displayed on the screen. The coincident pulses were readily observed in either case, but no delay could be established. We estimate that the half-life of the intermediate state is shorter than 0.3 microsecond.

VI. CONCLUSIONS

In order to utilize the measured values of the gamma-ray energies for establishing the level diagram of the product nucleus, Ra^{226}, the following facts are relevant: (1) the 67.8-kev line represents a transition to the ground level, as it is much stronger than any others; a level of this approximate energy is also directly known from the structure of the alpha-spectrum; (2) the final state of the 142-kev line is the 67.8-kev level, as proved by the observed coincidences; (3) since the line at 255 kev did not show coincidences with any other line, it is believed to represent a transition to the ground state. On this basis, the level diagram represented in Fig. 3 is suggested.

Several of the combinations possible according to this diagram were searched for but not observed. They must occur with considerably lower intensities than the three reported lines. The present experiments definitely disprove a level diagram of the type suggested by Jarvis and Ross.[6] These authors considered the possibility of two transitions of approximately equal energies near 68 kev in cascade with each other. If this were the case, there should be coincidences between the 67.8-kev line and the L x-rays. Such coincidences were looked for and found to be absent.

Angular momentum values may be assigned to the two lowermost states. The ground state is expected to have zero angular momentum like the ground states of all even-even nuclei. The 67.8-kev radiation seems definitely due to electric quadrupole. Rosenblum and Valadares[5] reported approximate intensities of four electron conversion lines. The line designated by them as "A" and attributed to conversion in the L_I level probably represents the unresolved contributions of the L_I and L_{II} levels, while the line designated as "B" represents conversion in the L_{III} levels. The fact that the "A" and "B" lines have approximately equal intensities definitely indicates electric quadrupole rather than magnetic dipole radiation.[12] Higher multipolarities appear incompatible with the reported upper limit for the half-life, while electric dipole seems to be excluded by the high value of the conversion coefficient. Hence the first excited state may be attributed angular momentum 2, in agreement with the general rule of Goldhaber.

From Table I it appears that we observed about 0.13 transitions of 67.8 kev per disintegration. To obtain the total intensity of the transition, one must add the radiationless (Auger) transitions from the L levels, plus the conversion in higher electron shells. These additional terms are difficult to evaluate. However, our results seem compatible with the observations of Falk-Vairant and Teillac,[7] who reported 0.22 conversion electrons per distegration, and those of Rosenblum, Valadares, and Vial[8] who found 24 percent excitation of the 67.8-kev level. The total conversion coefficient seems definitely comprised between 20 and 24.

We are greatly indebted to Dr. D. C. Stewart of the Argonne National Laboratory for supplying the ionium source which made this work possible.

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