Gamma-Radiation from Pb^{206}

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Gamma-rays accompanying the electron capture decay of 6.4 day Bi^{206} and the alpha-decay of 138-day Po^{210} have been observed by measurement of internal conversion electrons and photo-electrons with a lens spectrometer. The bismuth activity showed resolved lines corresponding to gamma-rays of 182, 234, 260, 341, 396, 470, 505, 536, 590, 803, 880, 889, 1020, 1097, and 1720 kev. A thin Po^{210} source of 480-mc alpha-particle strength was found to emit gamma-rays of 800±6 kev energy which yielded K and L internal conversion lines in the ratio of 3.7±0.5 to 1. The 803-kev gamma-ray observed in both the Bi^{206} and Po^{210} activities is identified with a transition from the first excited state of Pb^{206} to the ground state.

I. INTRODUCTION

THE present investigation originated in connection with a study[1] of the 68-min isomer of Pb^{204} and its parent, 12-hr Bi^{204}. The bismuth was produced by cyclotron bombardment of lead according to the reaction $Pb^{204}(d,2n)Bi^{204}$. Studies of gamma-radiation from the Pb^{204} isomer, chemically separated from the Bi^{204} parent, were made with a lens spectrometer. Measurements were also attempted on sources of bismuth, chemically separated from targets of ordinary lead, in order to observe the spectrum of Bi^{204} in equilibrium with Pb^{204}. An intense and complex structure of lines was found, most of which decayed with a half-life of about 6 days. To clearly distinguish the 12-hour Bi^{204} lines from the other activity present, cyclotron targets enriched in Pb^{204} by a factor of 15 over normal lead were used. It was suspected that the background activity was that associated with the electron capture decay of 6.4-day Bi^{206} produced from the more abundant Pb^{206} isotope by the $Pb^{206}(d,2n)Bi^{206}$ reaction.[2]

Because of recent general interest in the isotopes of lead the 6-day activity has been investigated further with the aim of substantiating its assignment to Bi^{206} and studying transitions in the residual nucleus Pb^{206} both in this and related processes. No previous spectrometer measurements on Bi^{206} have been reported while absorption experiments have yielded a variety of results.[2]

Other processes known to lead to the same product nucleus are the β^- decay of 4.23-min Tl^{206} and the α-decay of 138-day Po^{210}. Facilities for investigating the former are not yet available here. However, the polonium problem seemed feasible and is of interest both on account of its relationship to Bi^{206} and because a well-defined spectrometer measurement had not been made. It was hoped that such a study might assist in the determination of the group structure of the Po^{210} alpha-particle spectrum.

The polonium spectrum has long been thought to consist of a single alpha-particle line at 5.3 Mev. Weak gamma-radiation was also known[3] to be associated with the decay of polonium to the extent of about 1 quantum per 10^5 alphas and having an energy of about 0.8 Mev as determined by absorption experiments. Bothe examined[4] the internal conversion lines from a 30-mc Po^{210} source by means of a semi-circular focusing spectrometer at 7 percent resolution. He reported gamma-ray energies of 202, (355), (433), 798, and 1068 kev, of which the 798 line was the most intense; those in parenthesis were considered doubtful. Later Siegbahn and Slätis measured[5] the photo-electrons from a lead converter in a lens spectrometer and found one gamma-ray having an energy of 0.773 Mev in approximate agreement with Bothe's most intense line. An 86-mc polonium source was used and the resolution was sufficient to show only the K-shell electron peak: In separate absorption experiments the energy was checked and no other components were detected. An absorption measurement by DeBenedetti and Kerner[6] gave a similar result, but Zajac, Broda, and Feather found[7] in addition to the 0.8-Mev component, soft gamma-ray lines of which the most intense was at 84 kev.

The presence of gamma-radiation in the decay of Po^{210} indicates weak branching to one or more excited states of Pb^{206} and suggests that alpha-particles of energy lower than the 5.3 Mev group should occur. Chang examined[8] this region with an alpha-particle spectrograph of high resolution and found 12 weak lines spaced about 100 kev apart. The existence of such lines with the observed intensities would have been difficult to explain theoretically. In an experimental check[9] Wadey failed to confirm this fine structure and attributed Chang's results to source backing effects. The most recent alpha-spectrograph study was made[10] by Wagner who discovered 30 groups of alpha-particles

* Under contract with the AEC.

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[9] W. G. Wadey, Phys. Rev. **74**, 1846 (1949).

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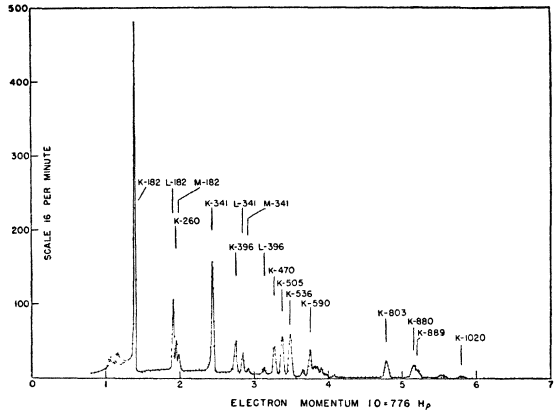

FIG. 1. Internal conversion spectrum of $Bi^{206}(K)Pb^{206}$ at 1.2 percent resolution.

from polonium samples. These were found to be independent of backing material and of intensity proportional to source thickness. It is stated, however, that the lines are probably not of nuclear origin, their explanation being unknown.

It appears, then, that a careful measurement of the polonium gamma-ray spectrum would serve as a guide to the location of the corresponding alpha-particle lines. In the present work this gamma-ray spectrum has been examined and compared with that of Bi^{206} decay.

II. EXPERIMENTAL

Gamma-ray measurements on bismuth and polonium activities were made with a lens spectrometer of conventional design. The instrument has a vacuum chamber 48 in. long and 10 in. i.d. and is equipped with a ring-focusing arrangement. With 5 mm diameter sources a resolution (full width at half maximum), of 1 percent can be attained. The coil current is regulated to about 0.1 percent by a circuit which compares the current shunt voltage with a potentiometer reference voltage. Calibration made with such standards as Cs^{137} or Co^{60} is generally reliable to about 0.5 percent.

Bi^{206} sources were prepared as follows: The deuteron-bombarded lead target was dissolved in hot 3 to 6N HNO_3, most of the lead removed as $PbCl_2$ by the addition of HCl, and the filtrate from the $PbCl_2$ made about 1N in HCl. This solution was then shaken for 20 to 30 minutes with a few hundred milligrams of nickel powder at 80 to 90°C in order to plate the carrier-free bismuth on the nickel. The nickel was centrifuged off, washed, and dissolved in hot 6N HNO_3, and 1 mg of iron as ferric salt was added to act as a carrier for the bismuth activity in the precipitation with excess ammonia which followed. To remove all the nickel, the ferric hydroxide (plus bismuth hydroxide) was washed, redissolved in acid, reprecipitated with ammonia, and washed again. It was then dissolved in 6N HCl, and the solution shaken with two successive portions of ethyl ether, previously equilibrated with 6N HCl. This removed the ferric ion almost quantitatively and left essentially carrier-free bismuth activity in the aqueous phase. The aqueous solution was heated to fuming with 1 ml of 6N H_2SO_4 to remove all chloride ions, and was then diluted to be about 2N in H_2SO_4. From this solution, the bismuth activity was deposited electrolytically. A small glass cell whose inside diameter at the bottom equalled the desired source diameter (usually 5 mm), was gasketed to a 0.001-in. thick copper foil which served as the cathode. A platinum stirrer was used as the anode and electrolysis was carried on for 30 to 90 minutes at a potential of about 3.5 volts. When no bismuth carrier was used the electrolysis yields were variable between 10 and 80 percent. Addition of 10 to 100 μg of bismuth carrier helped somewhat in assuring higher yields.

Observations were made on the 6-day bismuth spectrum after the 12-hour Bi^{204} lines had died out by measuring internal conversion lines in the lens spectrometer. Photo-electrons produced by Bi gamma-rays in a 21.5 mg/cm^2 lead converter were also examined. In this case a 2-mm thick brass plate separated the converter from the source in order to absorb the internal conversion electrons.

The Po^{210} source was specially prepared for this work by the AEC. It consisted of carrier-free polonium deposited on a nickel disk over an area 0.5 cm in diameter, and covered with 0.2 mil gold foil ($\sim$10 mg/cm^2) for the purpose of reducing the spread of contamination. The foil had been clamped with a metal ring and sealed at the outer edges with a vacuum evaporated coating. At the time of frabrication the alpha-particle strength was 478 mc, and from this the thickness of the source is estimated to be $\sim$0.5 mg/cm^2. Both the polonium internal conversion electrons and photo-electrons produced in a uranium converter by polonium gamma-rays were measured in the lens spectrometer.

III. EXPERIMENTAL RESULTS

Figure 1 shows the Bi^{206} internal conversion electron spectrum up to about 1.3 Mev taken at a resolution setting of 1.2 percent. Assignment of many of the resolved lines has been made as indicated in the figure. Not shown are weak K lines corresponding to 234- and 1097-kev gamma-rays. In addition, the internal conversion lines of a 1720 kev gamma-ray were observed after der Mateosian and Goldhaber[11] had shown that Bi^{206} produced photo-neutrons in beryllium but not in heavy water. The absence of background between the conversion lines in Fig. 1, other than that characteristic of scattering, indicates a K-capture process and no appreciable positron emission.

In the spectrum of photo-electrons due to Bi^{206} gamma-rays, many of the lines were not resolved because of the effects of converter thickness and the presence of a background of Compton electrons. However, the 1720-kev gamma-ray was again observed and

[11] E. der Mateosian and H. Goldhaber, Phys. Rev. **78**, 326(A) (1950).

a search made for other high energy lines which might have been missed in the internal conversion spectrum.

The half-life determined from measurements on the K-182 and K-341 lines over a period of 12 days was found to be 6.2±0.5 days in good agreement with the value 6.4 days assigned[12] to the K-capture decay of Bi^{206} by Templeton, Howland, and Perlman. All of the other prominent lines appeared to decay at this rate since their intensities relative to the K-182 line remained the same throughout the measurements. However the possibility that the weak 234- and 1097-kev lines decay at a somewhat different rate cannot be ruled out. The only other known activity of comparable half-life[13] which might be present is 14-day Bi^{205}.

Unresolved lines due to nuclear gamma-rays are still present in the internal conversion spectrum of Bi^{206} but intensities of sources were not sufficient to make the measurements at significantly better resolution (i.e. ~0.7 percent). The region between 50 and 80 kev shows a distribution which might well be Auger electrons due to x-rays following K-capture. This unresolved yield is indicated in Fig. 1 at a regulator dial setting in the vicinity of 1.2. Exposure of a Bi^{206} source in a 180° focusing spectrograph with a 100 gauss permanent magnet showed 6 resolved Auger lines between 55 and 73 kev. In addition, the K, L, M and N lines of the 182-kev gamma-ray and the K lines of 260- and 339-kev gamma-rays were observed in this instrument whose range is limited to below 300 kev.

Coincidence measurements carried out by A. W. Sunyar showed a rather large number of $\gamma\gamma$-coincidences from Bi^{206} sources, which is consistent with the very complex gamma-ray spectrum observed. But attempts to find delayed $\gamma\gamma$- or $X\gamma$-coincidences failed, which indicates that none of the excited states of Pb^{206} reached in the Bi^{206} decay has a half-life between 10^{-7} and 10^{-4} second. To establish whether any of these levels have life times above 1 second, rapid separations of lead from

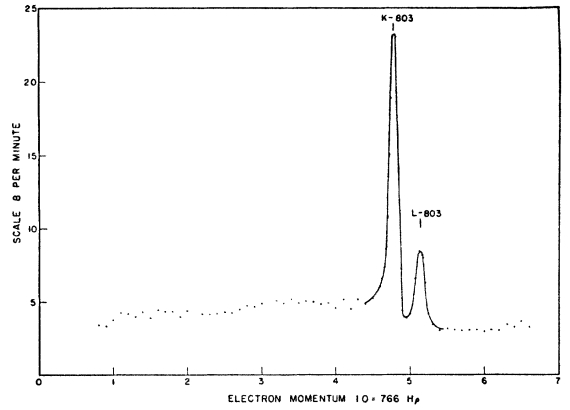

FIG. 2. Internal conversion electrons from a carrier-free Po^{210} source of 478 mc alpha-particle strength.

[12] Templeton, Howland, and Perlman, Phys. Rev. 72, 766 (1947).
[13] Way, Fano, Scott, and Thew, Nat. Bur. Standards, Circular 499.

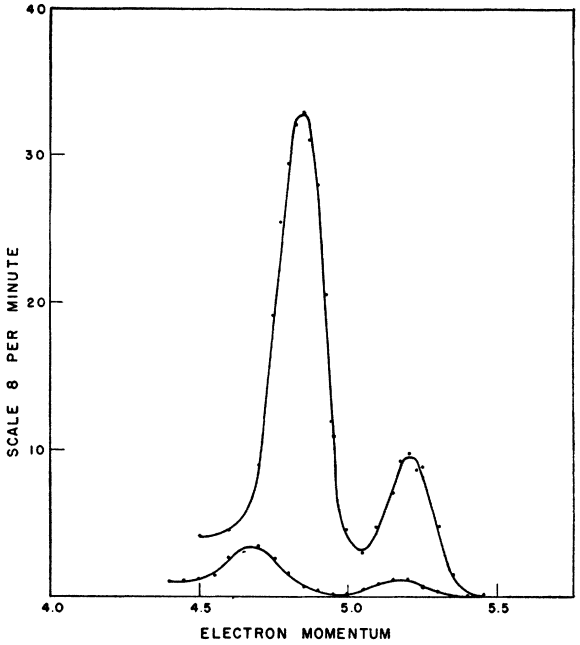

FIG. 3. A comparison of Po^{210} internal conversion electrons (upper curve) with photo-electrons from a 44 mg/cm² uranium converter (lower curve) both at 4.0 percent resolution.

Bi^{206} were carried out as follows. A small volume of hot 1N HCl solution of carrier-free Bi^{206} containing some Pb^{++} carrier was pulled through a bed of 200 mesh nickel powder placed on a sintered-glass funnel. Under the suction of flask receiving the filtrate was a G-M counter. The nickel removed 90 to 95 percent of the Bi, and the lead fraction could be counted within 1 to 2 seconds after separation. No active Pb^{206} of half-life greater than 1 second was found.

The Po^{210} internal conversion spectrum is given in Fig. 2 and shows that one gamma-ray is present yielding K and L-shell electrons in the ratio of 3.7±0.5 to 1. The gamma-ray energy is computed to be 800±6 kev, when a 10-kev correction has been made for energy loss in the gold foil protecting the source. The uncertainty of this correction is probably several kev because the foil thickness is not accurately known. The conversion lines here are somewhat asymmetrical due to scattering and absorption effects in both the source and protecting foil. This results in a line width of 2.5 percent although the spectrometer resolution in this case was 1.9 percent.

To study the Po^{210} gamma-ray spectrum further photo-electrons from uranium were examined at 4.0 percent resolution. The internally converted electrons were removed by a brass plate to which the uranium converter was attached and the arrangement was such that about 30 percent of all gamma-rays from the source were intercepted by the converter. The resulting photo-electron distribution is shown in the lower curve of Fig. 3 and may be compared with the internal conversion lines in the upper curve measured at the same spectrometer resolution setting. The fact that the

photo-electron lines are at somewhat lower energy is accounted for by the combined effects of energy reduction due to converter thickness (amounting to an average of ~23 kev) and the larger binding energy of uranium which for the K-shell is about 28 kev greater than that of lead.

A search was made for other gamma-rays from polonium both in the internal and external conversion studies covering a range of energies from 100 kev to 4 Mev. Figure 2 shows that up to 1.3 Mev there is no other conversion line of intensity greater than 5 percent of that of the K line of the 800-kev gamma-ray. The search at higher energies with the uranium converter gave negative results although, in this case, because of the rapid decrease of photoelectric yield with energy and the already small counting rate due to the 800-kev line, gamma-rays of moderate relative intensity might have been unobservable above the background.

IV. DISCUSSION

The uncertainty in energy determination of the bismuth lines is estimated to be about 0.5 percent. Since the 800-kev polonium gamma-ray is equal, within the probable error, to the 803-kev line is bismuth decay, it is reasonable to assume that both are, in fact, the result of the same nuclear transition. The bismuth value is considered to be the more accurate because of better resolution and the thinner sources used. The appearance of this gamma-ray in both spectra, taken together with the bismuth half-life measurement and lack of an electron continuum identifies the bismuth lines with the $Bi^{206}(K)Pb^{206}$ process.

Further evidence for the identity of the 803- and 800-kev gamma-rays might be obtained if a comparison of K/L ratios and internal conversion coefficients were possible. A measurement of the K/L ratio in the bismuth case could not be made because the L line is masked by the K lines of 880- and 889-kev gamma-rays. A determination of the internal conversion coefficient of the 803-kev bismuth line in the presence of so many other gamma-rays has not been feasible. In the polonium case an attempt to measure the conversion coefficient directly was made by W. Orr, but because of the very unfavorable source geometry, the results are at present inconclusive. However, with the following considerations a rough estimate of this coefficient may be obtained from the spectrometer curves of Fig. 3 in which the ratio of internal to external conversion is about 10. The yield of photo-electrons should be equal to the product of the gamma-ray strength, the efficiency of photoelectric conversion in the uranium converter including the solid angle it subtends at the source, a correction factor for the angular distribution of the photo-electrons, and the solid angle of the spectrometer. On the other hand, the internal conversion is isotropic so that in this case the yield is equal to the product of the gamma-ray strength, the conversion coefficient, and the spectrometer solid angle. In taking the ratio of these quantities the spectrometer solid angle and gamma-ray strength cancel giving the conversion coefficient in terms of the yield ratio and other factors which may be calculated. In this way, the conversion coefficient is found to lie between 0.01 and 0.05. This estimate has rather wide limits because the correction for the angular distribution of the photo-electrons is difficult to make under conditions in which the converter subtends a large solid angle at the source. Nothing definite can be said about the multipole order of the radiation since this range of values includes transitions from quadrupole to 2^4 pole according to the tables[14] of Rose, *et al.* Likewise, the measured K/L ratio cannot uniquely assign the multipole order since theoretical calculations at high Z have not as yet been made.

The absence of other lines from polonium suggests that the 803-kev gamma-ray is due to transition from the first excited state of Pb^{206} to the ground state. If lower excited states existed the barrier penetration probability for alpha-particle transitions to such states would be expected to be much greater than that for the transition to the 803-kev state because of the strong dependence of this probability on energy. Lower energy gamma-rays would therefore appear. States less than 100 kev above the ground state cannot be excluded since gamma-rays of such energy would not have been observable because of absorption of the conversion lines in the protective gold foil. The assignment of the 803-kev transition is in agreement with recent work by Harvey[15] in which the $Pb^{207}(d,t)Pb^{206}$ reaction was studied. Triton groups were observed corresponding to excited states in Pb^{206} at 0.86, 1.37, 1.71, 2.22, and 3.03 Mev. The curves[16] indicate a measuring accuracy of ~0.1 Mev; it is then possible that Harvey's first excited state corresponds to the 803-kev line found in the present work.

The remaining gamma-rays from Bi^{206} are presumably due to transitions from higher lying levels to the ground state, or between higher levels; several among these may be correlated with Harvey's data. However, the present resolution of the triton group structure is not sufficient to justify such comparisons and it is felt that a complete level scheme cannot be suggested at the present time. Further information would be obtained by resolving all of the bismuth gamma-rays and employing coincidence techniques to sort the various transitions. It is also hoped that a study of Tl^{206} beta-decay will help in establishing the levels in Pb^{206}.

We wish to thank Dr. M. Goldhaber for many useful suggestions in connection with this work. We are indebted to Mr. J. Bulkley and the crew of the MIT cyclotron for a number of lead irradiations and to Miss Elizabeth Wilson who gave valuable help in some of the source preparations and in the work with the 180° spectrograph.

[14] Rose, Goertzal, Spinrad, Harr, and Strong, private distribution.
[15] J. A. Harvey, MIT Prog. Report (April 1, 1950) (unpublished).
[16] J. A. Harvey (private communication to M. Goldhaber).